#### DESCRIPTION

#### SKIN TREATMENT COMPOSITION FOR WRINKLE REDUCTION

5 TECHNICAL FIELD
[0001]

The present invention relates to a skin treatment composition for wrinkle reduction. More specifically, it relates to a skin treatment

10 composition that reduces skin wrinkles by means of a film formed as said skin treatment composition dries after being applied on the skin.

#### BACKGROUND ART

15 [0002]

Conventionally, film agents have been used for a skin treatment composition for the purpose of reducing skin wrinkles. A film agent is used because the film shrinks as it dries; the idea is to pull small skin wrinkles with the film-forming contractile force to increase the tension and thus temporarily remove the small wrinkles. For this reason, film agents that have a strong contractile force and form a hard film have been used.

**25** [0003]

Known film agents that are blended into a wrinkle reducing agent using such strongly contracting polymers include, for example, polyurethane (see Patent Document 1); also, research has been done to study wrinkle reduction 5 by means of formation of a film having a strong contractile force by using film-forming polymers such as acrylic resins, vinyl acetate resins, polyethylene resins, silicone resins, polyvinyl 10 resins, polyvinyl alcohol, acrylic water soluble resins, cellulose water soluble resins, starch and its derivatives, gelatin, and sodium alginate (see Patent Document 2, for example). [0004]

contractile force is used, there are problems in that the wrinkle reduction effect is very limited and usability and the sensation during use are poor; this is because as the film agent contracts its adhesion to the skin weakens and it peels off easily from the skin, which is more elastic, and also because hardness of the film causes discomfort during use, premature breakage of the film, and glossy skin.

**25** [0005]

Patent Document 1: Japanese Patent Laid-Open H11-504949 bulletin

Patent Document 2: Japanese Patent Laid-Open H5-933 bulletin

5

DISCLOSURE OF INVENTION

[Problem that the present invention aims to solve]

The present invention has been carried out

10 in view of the situation described above, and its object is to provide a skin treatment composition for wrinkle reduction that is superior in terms of the wrinkle reduction effect, usability, and the sensation during use.

15 [Means to solve the Problem]
[0007]

[Invention of claims 1-12]

The inventors conducted earnest research and discovered that the aforementioned problems can be solved by preparing a skin treatment composition by blending a water dispersion of a specific polymer, specifically polyurethane and an acrylic type polymer, that forms a film having a weak contractile force, thus completing the present invention.

[8000]

The present invention is not based on the idea of using a conventional contracting film; on the contrary, it is based on the idea of 'filling's wrinkles with film.' That is, polyurethane and acrylic type polymers are used to obtain a new skin treatment composition for wrinkle reduction having superior functions than those conventionally known using an action mechanism different from those conventionally known: the skin treatment composition for wrinkle reduction of the present invention is applied on the skin to form a film on the skin that does not contract or become hard like the conventional ones.

15 [0009]

That is, the present invention is a skin treatment composition having a water dispersion of a polymer in which a non-water soluble film-forming polymer is dispersed in water wherein the main ingredients of said film-forming polymer are polyurethane having a film shrinkage rate of 20% or less and an acrylic type polymer having a film shrinkage rate of 20% or less.

[0010]

25 Said film-forming polymer is a polymer that

forms a film on the skin or substrate when a water dispersion of it is applied on the skin or substrate and dried.

[0011]

Said polyurethane is preferably polyurethane obtained by reacting an isocyanate compound and a diol compound containing a polyether diol, polycarbonate diol, and alkylene diol containing a carboxyl group; for said isocyanate compound it is preferable to contain isophorone diisocyanate.

[0012]

Furthermore, in said preferable polyurethane, the polyether diol is preferably polytetramethylene glycol, the polycarbonate diol is preferably polyhexamethylene carbonate diol, and the alkylene diol containing a carboxyl group is preferably dimethylolpropionic acid and/or dimethylolbutanoic acid.

Said acrylic type polymer is preferably a polymer of monomers whose main ingredient is ethyl acrylate.

[0014]

[0013]

15

Furthermore, it is preferable for the water

25 dispersion of the acrylic type polymer to contain

sulfonated polyvinyl alcohol.
[0015]

5

10

15

Also, in the present invention, the film strength of said polyurethane is preferably  $300-700 \text{ kg/cm}^2$ , and the film strength of the acrylic type polymer is preferably  $0.1-100 \text{ kg/cm}^2$ .

The film elongation of the polyurethane is preferably 200-500%, and the film elongation of the acrylic type polymer is preferably 500-2000%.

[0017]

Also, the average particle size of the polyurethane in said water dispersion of polyurethane is preferably 10-300 nm, and the average particle size of the acrylic type polymer in said water dispersion of acrylic type polymer is preferably 100-600 nm.

[0018]

In the present invention, the polyurethane

20 in the water dispersion of polyurethane preferably
is a mixture of particles having an average
particle size of 20-60 nm and particles having an
average particle size of 150-200 nm.

[0019]

25 The skin treatment composition for wrinkle

reduction of the present invention preferably contains the polyurethane to 1-10 wt% of the total weight of the skin treatment composition and the acrylic type polymer to 1-20 wt% of the total weight of the skin treatment composition.

[0020]

5

The present invention can also contain scaly silica. By adding scaly silica, the film formed by the water dispersion of the polymer becomes

10 thicker and the wrinkle reduction effect is augmented and reinforced and thus a skin treatment composition for wrinkle reduction that can accommodate the elasticity and movement of the skin for a long duration of time can be obtained.

15 [0021]

The contractile force in the present invention is a force with which the water dispersion of the polymer contracts when it forms a film, and this force is evaluated based on the film shrinkage rate. The film shrinkage rate is a degree of contraction of the film relative to the thin layer of the original water dispersion when said thin layer of the polymer water dispersion is dried to form the film. Specifically, a water

polymer is poured into a 5 cm x 5 cm polyethylene mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature  $(50^{\circ}\text{C})$  for a prescribed amount of time (three days at room temperature) to obtain a film, and the vertical height and horizontal length of this film are measured and used in the following formula for calculation.

[0022]

5

Film shrinkage rate (%) = [(Vertical measurement x Horizontal measurement)/25] x 100

The film strength and the film elongation of the polymer is the strength and elongation of the 15 film obtained by drying the thin layer of the water dispersion of the polymer; specifically, a water dispersion of a solid equivalent 1 g of the polymer is poured into a 5 cm x 5 cm polyethylene mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature 20 (50°C) for a prescribed amount of time (three days at room temperature) to obtain a film, and this film is cut out using a dumbbell #3 and the autograph function of "Tensile tester RTM-250" 25 from Orientec Co., Ltd. is used to carry out the

[0024]

[Invention of claims 13-25]

The inventors conducted earnest research to solve the aforementioned problems and discovered that the aforementioned problems can be solved by preparing a skin treatment composition by blending in a non-emulsifying type cross-linked silicone and polyurethane that forms a film having a weak contractile force, thus completing the present invention.

[0025]

The present invention is not based on the

idea of using a conventional contracting film; on
the contrary, it is based on the idea of using a
paste-like material that can change its form
freely according to the wrinkle site and noncontracting film, thus 'filling wrinkles with

film'. That is, a non-emulsifying type free-form
cross-linked silicone and polyurethane, as a noncontracting film agent, are used to obtain a new
skin treatment composition for wrinkle reduction
having superior functions than those

conventionally known using an action mechanism

different from those conventionally known. [0026]

That is, the present invention is a skin treatment composition for wrinkle reduction

5 comprising (a) a non-emulsification type cross-linked silicone, (b) a film forming polymer having a film shrinkage rate of 20% or less containing as a main ingredient a polyurethane having a film shrinkage rate of 20%, (c) a liquid oil component, and (d) water.

[0027]

Said film-forming polymer is a polymer that forms a film on the skin or substrate when a water dispersion of it is applied on the skin or substrate and dried.

[0028]

15

Said non-emulsification type cross-linked silicone is preferably one, two, or more chosen from a group consisting of a cross polymer derived from a reaction between methyl hydrogen polysiloxane and methyl vinyl polysiloxane, a cross polymer derived from a reaction between a partial long chain alkylated methyl hydrogen polysiloxane and methyl vinyl polysiloxane, and a cross polymer derived from a reaction between

methyl hydrogen polysiloxane and alkene.
[0029]

The long chain alkyl of said partial long chain aklylated methyl hydrogen polysiloxane should preferably have 10-14 carbon atoms.

The non-emulsifying type cross-linked silicone should preferably be added as it is swollen with the liquid oil component.

10 [0031]

5

Said polyurethane is preferably polyurethane obtained by reacting an isocyanate compound and a diol compound containing a polyether diol, polycarbonate diol, and alkylene diol containing a carboxyl group; said isocyanate compound preferably contains isophorone diisocyanate.

[0032]

Furthermore, in said preferable polyurethane, the polyether diol is preferably

20 polytetramethylene glycol, the polycarbonate diol is preferably polyhexamethylene carbonate diol, and the alkylene diol containing a carboxyl group is preferably dimethylolpropionic acid and/or dimethylolbutanoic acid.

**25** [0033]

Also, in the present invention, the film strength of said polyurethane is preferably  $300-700\ kg/cm^2$ .

[0034]

5 The film elongation of the polyurethane is  $preferably \ 200-500\%.$ 

[0035]

Said film forming polymer having a film shrinkage rate of 20% or less containing as a main ingredient a polyurethane having a film shrinkage rate of 20% is preferably added in the form of a water dispersion.

[0036]

10

The average particle size of the

15 polyurethane in said water dispersion of polyurethane is preferably 10-300 nm.

[0037]

In the present invention, the polyurethane in said water dispersion of polyurethane is

20 preferably a mixture of particles having an average particle size of 20-60 nm and particles having an average particle size of 150-200 nm.

[0038]

The skin treatment composition for wrinkle reduction of the present invention preferably

contains the non-emulsifying type cross-linked silicone to 0.5-5.0 wt% of the total amount of the skin treatment composition, and the polyurethane having a film shrinkage rate of 20% or less to 0.1-10.0 wt% of the total amount of the skin treatment composition.

[0039]

5

The contractile force in the present invention is a force with which the water 10 dispersion of the polymer contracts when it forms a film, and this force is evaluated based on the film shrinkage rate. The film shrinkage rate is a degree of contraction of the film relative to the thin layer of the original water dispersion when 15 said thin layer of the polymer water dispersion is dried to form the film. Specifically, a water dispersion of a solid equivalent 1 g of the polymer is poured into a 5 cm x 5 cm polyethylene mold such that the film thickness is approximately 20 0.5 mm and dried at a prescribed temperature (50°C) for a prescribed amount of time (three days at room temperature) to obtain a film, and the vertical height and horizontal length of this film are measured and used in the following formula for 25 calculation.

[0040]

Film shrinkage rate (%) = [(Vertical measurement x Horizontal measurement)/25] x 100

- The film strength and the film elongation of 5 the polymer is a strength and elongation of the film obtained by drying a thin layer of the water dispersion of the polymer; specifically, a water dispersion of a solid equivalent 1 g of the polymer is poured into a 5 cm x 5 cm polyethylene 10 mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature  $(50\,^\circ\!\!\mathrm{C}\,)$  for a prescribed amount of time (three days at room temperature) to obtain a film, and this 15 film is cut out using a dumbbell #3 and the autograph function of "Tensile tester RTM-250" from Orientec Co., Ltd. is used to carry out the measurement at 20% and a cross head speed of 300mm/min.
- 20 [Effects of the invention]
  [0042]

25

The skin treatment composition for wrinkle reduction of the present invention is not sticky and at the same time soft, supple, elastic, and flexible; it does not crack or peel off; it

adheres well to the skin and gives a light tactile sensation without discomfort; it can be worn for a long duration of time with ease and maintain a state of removing even small wrinkles for a long duration of time (i.e. the wrinkle reduction effect lasts for a long duration of time); it therefore exhibits a wrinkle reduction effect that hasn't been known conventionally.

Furthermore, this effect is augmented and reinforced by adding scaly silica, resulting in an exceptional wrinkle reduction effect. In particular, the film formed by the water dispersion of the polymer becomes thick and the wrinkle reduction effect is augmented and reinforced and thus a skin treatment composition for wrinkle reduction that can accommodate the elasticity and movement of the skin for a long duration of time can be obtained.

20

5

BEST MODE FOR CARRYING OUT THE INVENTION
[0044]

[Invention of claims 1-12]

The best embodiments of the present invention are described in detail below.

[0045]

5

10

The skin treatment composition for wrinkle reduction of the present invention contains a water dispersion of a polymer in which a water-insoluble film forming polymer is dispersed in water, and the main ingredients of said film forming polymer are polyurethane having a film shrinkage rate of 20% or less and an acrylic type polymer having a film shrinkage rate of 20% or less. The film shrinkage rate is defined as described above.

[0046]

Those for which said film shrinkage rate is over 20% have a poor wrinkle reduction effect, 15 peel off the skin, cause noticeable "gloss", and exhibit poor usability due to discomfort; therefore they cannot manifest the effects of the present invention. The preferable range of the film shrinkage rate is 10% or less. The lower 20 limit of the film shrinkage rate is 0, i.e. no shrinkage, which is preferable; however, 5% is sufficient and also practical. That is, the film shrinkage rate in the present invention is 0-20%; within this range, preferable combinations of the 25 upper and lower limits are used, examples include 5-20%, 0-10%, and 5-10%. [0047]

In the present invention, as described above, a water dispersion of polyurethane, in which nonwater-soluble polyurethane is dispersed in water, 5 is used; said water dispersion is applied on the skin or substrate and as it is dried a polyurethane film is formed on the skin or substrate.

#### [0048] 10

The polyurethane used in the present invention is a polymer having urethane bonds; the urethane bond is formed by an addition reaction between an isocyanate group and a compound having 15 active hydrogen, such as a hydroxyl group. The polyurethane in the present invention is preferably obtained with a conventional method reacting at least (A) an isocyanate compound having two isocyanate groups and (B) a diol 20 compound having two hydroxyl groups.

[0049]

25

Selection of said isocyanate compound (ingredient A) is not limited in particular as long as it is used in conventional polyurethane manufacturing, examples include organic

diisocyanate compounds such as aliphatic
diisocyanate compounds, alicyclic isocyanate
compounds, and aromatic diisocyanate compounds.

More preferable are aliphatic diisocyanate
compounds and alicyclic diisocyanate compounds.

One, two or more isocyanate compounds are freely
selected and used.

## [0050]

5

Examples of said aliphatic diisocyanate

10 compounds include ethylene diisocyanate, 2,2,4trimethylhexamethylene diisocyanate, and 1,6hexamethylene diisocyanate.

[0051]

Examples of said alicyclic diisocyanate

15 compounds include hydrogenated 4,4'diphenylmethane diisocyanate, 1,4-cyclohexane
diisocyanate, methylcyclohexylene diisocyanate,
isophorone diisocyanate (hereafter referred to as
"IPDI"), and norbornane diisocyanate.

# 20 [0052]

Examples of said aromatic diisocyanate compound include 4,4'-diphenylmethane diisocyanate, xylylene diisocyanate, toluene diisocyanate, and naphthalene diisocyanate.

#### **25** [0053]

Of the specific examples of said isocyanate compound (ingredient A), 1,6-hexymethylene diisocyanate, IPDI, and norbornane diisocyanate are preferable due to superior weather resistance and availability. IPDI is particularly preferable. [0054]

Selection of said diol compound (ingredient B) is not limited in particular as long as it is used in conventional polyurethane manufacturing,

10 preferable examples include an alkylene diol, carboxyl group-containing alkyl diol, alicyclic diol, spiro diol, polyester diol, polyether diol, polycarbonate diol, polybutadiene diol, polyisoprene diol, and polyolefin diol. Of these, particularly preferably used are an alkylene diol, carboxyl group-containing alkylene diol, alicyclic diol, polyether diol, and polycarbonate diol. One two or more diol compounds are freely selected and used.

## 20 [0055]

5

Examples of said alkylene diol include ethylene glycol, propylene glycol, 1,4-butane diol, 1,6-hexane diol, neopentyl glycol, 1,8-octane diol, and 1,10-decane diol.

#### **25** [0056]

Preferable examples of said carboxyl groupcontaining alkylene diol include carboxylic acids
having 3-26 carbons, more preferably 3-12 carbons,
as well as having a dialylol groups, such as
dimethylol, diethanol and propanol. Specific
examples include dimethylol propionic acid
(hereafter "DMPA") and dimethylol butanoic acid
(hereafter "DMBA"); a mixture thereof can be used
as well.

# 10 [0057]

Examples of said alicyclic diol include 1,4-cyclohexane dimethanol (hereafter "CHDM"), which is preferable because it gives adequate strength to the film.

# 15 [0058]

Preferable examples of said spiro diol include spiro glycol.

[0059]

Examples of said polyester diol include

those obtained by condensation polymerization

between at least one chosen from a group of

dicarboxylic acids including succinic acid,

glutaric acid, adipic acid, sebacic acid, azelaic

acid, maleic acid, fumaric acid, phthalic acid,

and terephthalic acid and at least one chosen from

a group of diols including ethylene glycol, propylene glycol, 1,4-butane diol, 1,6-hexane diol, neopentyl glycol, 1,8-octane diol, 1,10-decane diol, diethylene glycol, polyethylene glycol

5 (hereafter "PEG"), polypropylene glycol (hereafter "PPG"), tetramethylene glycol, polytetramethylene glycol, and spiro glycol, as well as those obtained by the ring opening polymerization of lactonic acid.

# 10 [0060]

Examples of said polyether diol include

polyether diol contained in diol used in synthesis

of said polyester diol, examples of which include

a polyoxyethylene diol such as diethylene glycol

15 and PEG, a polyoxypropylene diol such as PPG, a

polyoxytetramethylene diol such as

polytetramethylene glycol (hereafter "PTMG"),

phenols such as bisphenol A, and a product of the

ring opening addition polymerization of bisphenol

20 A and at least one compound chosen from propylene

oxide (hereafter "PO") and ethylene oxide

(hereafter "EO") (if a copolymer is used, such a

copolymer can be either a block copolymer or

25 are preferable, and PTMG is particularly

random copolymer). Of these, PEG, PPG, and PTMG

preferable since it forms a soft film and adheres to the skin very well.
[0061]

Preferable examples of said polycarbonate

5 diol include polyhexamethylene carbonate diol
(hereafter "PHMC") because of its ability to form
a soft film and its superior adhesion to the skin.
[0062]

In the present invention, polyurethane that

10 forms a superior film can be obtained by using the diol compound (ingredient B) consisting of a mixture of a polyether diol, polycarbonate diol, and carboxyl group-containing alkylene diol.

[0063]

In the present invention, when a carboxyl group-containing alkylene diol is used for the diol compound in the synthesis to obtain polyurethane containing carboxyl groups in the molecule, the carboxyl groups incorporated in the 20 molecule can be neutralized by neutralizers such as triethylamine, trimethylamine, 2-amino-2-methyl-1-propanol, triethanolamine, potassium hydroxide, and sodium hydroxide to obtain a stable water dispersion of polyurethane.

25 [0064]

In the present invention, when mixing a polyether diol, polycarbonate diol, and carboxyl group-containing alkylene diol, it is particularly preferable to use PTMG for said polyether diol, PHMC for the polycarbonate diol, and DMPA and/or

DMBA for said carboxyl group-containing alkylene diol.

[0065]

5

In the present invention, when using these

10 preferable diol compounds, it is particularly preferable to use an isocyanate compound that contains IPDI; a skin treatment composition for wrinkle reduction having an exceptional wrinkle reduction effect can be obtained by using

15 polyurethane synthesized by using IPDI for the isocyanate and PTMG, PHMC, DMPA and/or DMBA for the diol compounds.

[0066]

The molar ratio of said isocyanate compound (ingredient A) and diol compound (ingredient B) is preferably A/B = 2/0.8-2/1.8, and more preferably A/B = 2/1-2/1.8.

[0067]

In the present invention, it is preferable

25 to use polyurethane having structural units

derived from alkylene oxide (RO); this makes it easier to control the elongation of the film obtained from the water dispersion of the polyurethane to obtain a flexible film. A skin treatment composition that manifests superior effects and superior usability can be obtained by using this polyurethane for the preparation.

[0068]

5

25

Examples of the compound having a structural 10 unit derived from RO include a polyoxyethylene diol such as diethylene glycol and PEG, a polyoxypropylene diol such as PPG, a polyoxytetramethylene diol such as PTMG, polyoxyethylene polyoxypropylene glycol (EO/PO 15 block copolymer), phenols such as bisphenol A, and a product of the ring opening addition polymerization of bisphenol A and at least one compound chosen from PO and EO (if a copolymer is used, such a copolymer can be either a block 20 copolymer or random copolymer); preferably used are PEG, PPG, PTMG, etc. [0069]

In the present invention, the compound having a structural unit derived from RO is used as a polyether diol ingredient in said ingredient

Β.

[0070]

The water dispersion of polyurethane is prepared with a conventional method; for example,

5 a pre-polymer having remaining isocyanate obtained from a reaction in an organic solvent is dispersed in water containing potassium hydroxide under high speed agitation, followed by a chain elongation reaction to increase the molecular weight, and

10 said organic solvent is recovered from the obtained water based solution to obtain the water dispersion of the polyurethane.

[0071]

In the present invention, the following is

15 particularly preferable for said polyurethane and
water dispersion of polyurethane. That is:

[0072]

With regard to the film characteristics of the polyurethane, the strength is 300-700 kg/cm², 20 and preferably 400-600 kg/cm². If the strength is less than 300 kg/cm², then the film cannot follow the skin movements and peeling easily occurs. If it is over 700 kg/cm², then the film causes discomfort on the skin. The elongation is 200-25 500%, preferably 300-500%. If the elongation is

less than 200%, then the film formed on the skin cannot follow the skin movements very well. If it is over 500%, then the wrinkle reduction effect becomes insufficient.

## 5 [0073]

The average particle size of the polyurethane in the water dispersion of the polyurethane is 10-300 nm, preferably 20-200 nm.

If the average particle size is less than 10 nm,

then there is a sufficient small-wrinkle reduction effect but on the other hand the large-wrinkle reduction effect becomes insufficient. If the average particle size is over 300 nm, then adhesion to the skin becomes poor and peeling tends to occur.

#### [0074]

A preferable embodiment in the present invention uses two kinds of water dispersions of polyurethane having different particle sizes.

20 This way, not only the wrinkle reduction effect but also a skin mark erasing effect can be obtained. For said two kinds of polyurethane having different particle sizes, polyurethanes having an average particle size of 20-60 nm and an average particle size of 150-200 nm are preferable.

[0075]

5

10

The blend ratio of the polyurethane is preferably 0.1-10 wt% of the total amount of the skin treatment composition. If the blend ratio is less than 0.1 wt% then the effect of the present invention cannot be obtained sufficiently; and if it is over 10 wt% then the film tends to peel off the skin. A more preferable blend ratio range is 1-8 wt% of the total amount of the skin treatment composition.

[0076]

Also, in the present invention, as described above, a water dispersion of an acrylic type polymer, i.e. a water-insoluble acrylic type 15 polymer is dispersed in water, is used; said water dispersion is applied on the skin or substrate and upon drying forms a acrylic type polymer film on the skin or substrate. For said water dispersion, it is preferable to use those conventionally known 20 as polymer emulsions such as a polymer emulsion obtained by emulsification polymerization of acrylic type monomers. Said acrylic type polymer has so-called film forming properties, which means, when the polymer emulsion is applied on the skin or substrate, it forms a film on the skin or 25

substrate as it dries. Therefore, although in the present invention the acrylic type polymer emulsion is preferably dispersed, i.e. diluted, in water before blending, it is also acceptable to use the acrylic type polymer emulsion as is for the water dispersion.

[0077]

5

For the acrylic type polymer of the present invention, polymers derived from monomers 10 containing acrylic ester type monomers such as acrylic ester and methacrylic ester are preferable. Specific examples of said acrylic ester type monomers include methyl acrylate, ethyl acrylate, propyl acrylate, n-butyl acrylate, tert-butyl 15 acrylate, hexyl acrylate, cyclohexyl acrylate, octyl acrylate, 2-ethylhexyl acrylate, methyl methacrylate, ethyl methacrylate, propyl methacrylate, n-butyl methacrylate, tert-butyl methacrylate, hexyl methacrylate, cyclohexyl 20 methacrylate, octyl methacrylate, and 2-ethylhexyl methacrylate. The acrylic ester type monomers are used individually or in combinations of two or more.

[0078]

25 The monomers for making said acrylic type

polymer can contain, in addition to the acrylic ester type monomers, other hydrophobic monomers. Examples of said hydrophobic monomers include aromatic mono- and di- vinyl compounds such as 5 styrene,  $\alpha$  -styrene, chlorostyrene, alkyl styrene, and divinyl styrene, vinyl cyanide compounds such as acrylonitrile and methacrylonitrile, vinyl esters such as vinyl acetate, vinyl halogenate such as vinyl chloride and vinylidene chloride, 10 fluorocarbon monomers such as trifluoroethyl methacrylate, 2, 2, 3, 3-tetrafluoropropyl methacrylate, 2, 2, 3, 3, 4, 4-hexafluorobutyl methacrylate, perfluorooctyl methacrylate, and perfluorooctyl acrylate, and silicone macro

[0079]

monomers.

15

20

25

It is common to copolymerize the acrylic type polymers of the present invention by further adding hydrophilic monomers. This way, stable polymer emulsion, i.e. stable water dispersion of an acrylic type polymer, can be obtained.

[0080]

Examples of the hydrophilic monomers include ethylene type unsaturated carboxylic acids such as acrylic acid, methacrylic acid, itaconic acid,

maleic acid, fumaric acid, and crotonic acid, ethylene type monomers containing a hydroxyl group such as hydroxyethyl acrylate, hydroxyethyl methacrylate, glycidyl acrylate, glycidyl

- 5 methacrylate, ethylene glycol diacrylate, ethylene glycol dimethacrylate, polyethylene glycol monoacrylate, and polyethylene glycol monomethacrylate, ethylene type amides such as N-methylol methacrylamide and N-diacetone acrylamide,
- aminoethyl methacrylate, N, N-dimethylaminoethyl acrylate, acrylate, N, N-dimethylaminoethyl acrylate, N, N-dimethylaminoethyl methacrylate, N, N-diethylaminoethyl acrylate, N, N-diethylaminoethyl acrylate, N, N-diethylaminoethyl methacrylate, N, N-
- trimethylaminoethyl acrylate, N,N,Ntrimethylaminoethyl methacrylate, as well as the salts thereof.

[0081]

If the hydrophilic monomers having a

20 carboxyl group in the structure are used for preparing the acrylic type polymer emulsion, a neutralizer can be used to neutralize the carboxyl groups incorporated in the molecule to achieve superior dispersion of the acrylic type polymer

25 into water. Examples of the neutralizer for said

carboxyl group include triethylamine, trimethylamine, 2-amino-2-methyl-1-propanol, triethanolamine, potassium hydroxide, and sodium hydroxide.

## 5 [0082]

The monomers containing acrylic type ester monomers for preparing the acrylic type polymer pertaining to the present invention can be used individually or in combinations of two or more kinds; a preferable combination has 70-100 mole% hydrophobic monomers and 0-30 mole% hydrophilic monomers, and a more preferable combination has 85-99 mole% hydrophobic monomers and 1-15 mole% hydrophilic monomers.

## **15** [0083]

10

For said acrylic type polymer of the present invention, a polymer polymerized from monomers whose main ingredient is an acrylic type ester monomer consisting of an acrylic ester and/or

20 methacrylic ester is preferable. More preferable is a polymer from monomers whose main ingredient is ethyl acrylate and/or ethyl methacrylate, and even more preferable is a polymer from monomers whose main ingredient is ethyl acrylate.

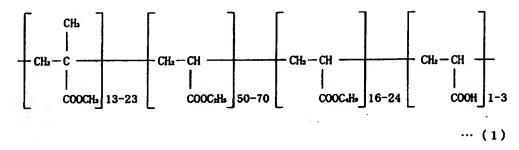
## **25** [0084]

If hydrophilic monomers are to be included, acrylic acid and/or methacrylic acid are preferable for said monomers.

Examples of a specific embodiment of the preferable acrylic type polymer in the present invention include the following formula (1):

[0086]

[Chemical formula 1-1]



[0087]

10

[8800]

The acrylic type polymer emulsion used in the present invention should preferably contain

15 sulfonated polyvinyl alcohol as an emulsifier/colloid agent to obtain finely textured film. It is particularly preferably to have this for said emulsification polymerization.

[0089]

20 Examples of preferably used sulfonated polyvinyl alcohol include the following formula

(2):

[0090]

[Chemical formula 1-2]

5 [0091]

Therefore, the water dispersion of the acrylic type polymer of the present invention should preferably contain sulfonated polyvinyl alcohol.

10 [0092]

Also, when preparing said acrylic type
polymer emulsion, it is preferable to add a
surfactant to stabilize dispersion at the time of
the emulsion polymerization. Selection of the

15 surfactant used is not limited in particular; a
common anionic, cationic, or non-ionic surfactant
can be used. It is also possible to use two or
more kinds, such as a combination of an anionic
type and non-ionic type and a combination of a

20 cationic type and non-ionic type. Of said
surfactants, a non-ionic surfactant is preferable.
[0093]

Examples of the non-ionic surfactant include

polyoxyethylene (hereafter "POE") alkyl ether, POE alkylphenyl ether, and POE-polypropylene oxide (hereafter "POP") block copolymer; examples of the anionic surfactant include alkylbenzene sulfonate, and alkylnaphthalene sulfonate, and polyethylene 5 oxide alkyl ether sulfate. Examples of the cationic surfactant include primary, secondary, and tertiary amine salts, and quaternary ammonium salts having an aliphatic hydrocarbon group. 10 preferable non-ionic surfactant is POE alkyl ether, of which even more preferable is an alkyl ether having 12-20 POE carbons; POE oleyl ether is particularly preferable. The POE addition mole number is 30-65 moles, and more preferable is 40-15 60 moles.

[0094]

The amount of the surfactant added is preferably 5 weight parts or less, more preferably 3 weight parts of less, per 100 weight parts of the monomers used. If it is over 5 weight parts then the physical properties of the film deteriorates.

[0095]

In the present invention, the following is

25 even more preferable for said acrylic type polymer

and the water dispersion of the acrylic polymer (acrylic polymer emulsion or water dispersion thereof). That is:

- For the film strength of the acrylic type 5 polymer, the strength is  $0.1-100 \text{ kg/cm}^2$ , more preferably  $10-70 \text{ kg/cm}^2$ . If the strength is less than 0.1 kg/cm<sup>2</sup>, then the film cannot follow the skin movements and peeling easily occurs. is over  $100 \text{ kg/cm}^2$ , then the film causes 10 discomfort on the skin. The elongation is 300-2000%, preferably 500-1000%. If the elongation is less than 300%, then the film formed on the skin cannot follow the skin movements very well. is over 2000%, then the wrinkle reduction effect 15 becomes insufficient. The glass transition temperature (Tg) of the acrylic type polymer is preferably 0°C or lower.
- The average particle size of the acrylic type polymer in the water dispersion of the acrylic type polymer is 100-600 nm. If the average particle size is less than 100 nm, then there is a sufficient small-wrinkle reduction effect but on the other hand the large-wrinkle

[0097]

reduction effect becomes insufficient. If the average particle size is over 600 nm, then adhesion to the skin becomes poor and peeling tends to occur.

## 5 [0098]

The blend ratio of the acrylic polymer is preferably 1-20 wt% of the total amount of the skin treatment composition. If the blend ratio is less than 1% then the effect of the present

10 invention cannot be obtained sufficiently; if it is over 20 wt% then the viscosity of the skin treatment composition become high and preparation of formulations and application on the skin become difficult. A more preferable blend ratio range is 5-15 wt% of the total amount of the skin treatment composition.

#### [0099]

The present invention uses said film forming polymers of the present invention as the main

20 ingredient and preferably consists of these polymers; however, other film forming polymers can be added within the range that does not affect the effect of the present invention, as long as the film shrinkage rate of the total film forming

25 polymer is 20% or less.

[0100]

The skin treatment composition of the present invention can further contain scaly silica; this reinforce the wrinkle reduction

5 effect of the film consisting of the polyurethane and acrylic type polymer as described above.

Specifically, the film formed by the water dispersion of the polymer becomes thick and the wrinkle reduction effect is augmented and

10 reinforced and thus a skin treatment composition for wrinkle reduction that can accommodate the elasticity and movement of the skin for a long duration of time can be obtained.

[0101]

15 Scaly silica is scale-like silica particles that have a film forming capability of their own and are able to form a strong film at ordinary temperatures. Scaly silica has a laminated particle configuration; it substantially consists 20 of leaf-like secondary particles formed by multiple thin flake primary particles stacked together with their planes oriented parallel to each other. In the present invention, it is

preferable to use micro-scaly silica.

**25** [0102]

Scaly silica is commercially available; examples of commercial products that can be used include Sunlovely LFS-C (from Dohkai Chemical Industries Co., Ltd.).

### 5 [0103]

10

In the present invention, a preferable blend ratio of scaly silica is 0.1-5.0 wt%, more preferably 0.5-3.0 wt%, of the total amount of the skin treatment composition. If the blend ratio is less than 0.1 wt%, then the effect of adding scaly silica cannot be manifested sufficiently; if it is over 5.0 wt% then whiteness is seen on the skin surface, which is visually less agreeable.

[0104]

In addition to the aforementioned ingredients, other ingredients used in skin treatment compositions such as cosmetics and drugs can be blended as necessary into the skin treatment composition of the present invention as long as the effect of the present invention is not adversely affected. The aforementioned optional ingredients include oil components, power ingredients other than those mentioned above, surfactants, humectants, water soluble polymers, thickeners, film forming agents other than those

of the present invention, ultraviolet absorbents, sequestering agents, sugars, amino acids, organic amines, pH adjustment agents, nutritional supplements for skin, vitamins, antioxidants, and perfumes.

[0105]

5

10

The skin treatment composition for wrinkle reduction of the present invention can be prepared by blending in the aforementioned ingredients and following a conventional method.

[0106]

The skin treatment composition of the present invention is used in foundation cosmetic formulations in the form of cream, emulsion, or

15 lotion.

[0107]

[Invention of claims 13-25]

The best embodiments of the present invention are described in detail below.

20 [0108]

The non-emulsifying type cross-linked silicone of the present invention is described in detail below.

[0109]

Non-emulsifying type cross-linked silicone

is a cross-linked silicone in which some of silicone chains are cross-linked, characterized by not having its own ability to emulsify oil and water. A cross-linked silicone is verified as 5 non-emulsifying when a composition having water, oil, and the cross-linked silicone is stirred at a high speed using a homomixer and as a result emulsification does not occur or emulsification occurs but the particle size of the emulsified 10 particles is large, 50 micrometers or more, and the emulsified state does not last when allowed to stand for a while.

[0110]

Examples of the non-emulsifying cross-linked

15 silicone used in the present invention include a cross polymer derived from a reaction between methyl hydrogen polysiloxane and methylvinyl polysiloxane (hereafter

"dimethicone/vinyldimethicone cross polymer"), a

20 cross polymer derived from a reaction between
partial long chain alkylated methyl hydrogen
polysiloxane and methylvinyl polysiloxane
(hereafter "vinyldimethicone/alkyldimethicone
cross polymer"), and a cross polymer derived from

25 a reaction between methyl hydrogen polysiloxane

and alkene (hereafter "dimethicone cross polymer").

In the present invention, it is preferable to use one, two or more types selected from a group of the aforementioned three types of cross polymers.

5 [0111]

For the aforementioned methylvinyl polysiloxane, those that have at least two vinyl groups in the molecule are used in the present invention to effectively produce cross polymers.

10 For this production, methylvinyl polysiloxane having one vinyl group in the molecule is commonly used and this controls the cross-link ratio of the cross-polymer.

[0112]

The number of carbons in the long chain alkyl in the partial long chain alkylated methyl hydrogen polysiloxane can be set at will; in this invention a preferable number is 10-14 and a lauryl group, having 12 carbons, is the most

[0113]

preferable.

20

25

For the aforementioned alkene, those that have at least two vinyl groups in the molecule are used in the present invention to effectively produce cross polymers. For this production,

alkene having one vinyl group in the molecule is commonly used and this controls the cross-link ratio of the cross-polymer.

[0114]

In the present invention, the aforementioned 5 dimethicone/vinyldimethicone cross polymer corresponds to INCI designation "dimethicone/vinyldimethicone cross polymer" or "polysilicone-11". Dimethicone cross polymer corresponds to INCI designation "dimethicone cross 10 polymer". Of vinyldimethicone/alkyldimethicone cross polymers, a cross polymer derived from a reaction between laurylated methyl hydrogen polysiloxane and methyl vinyl polysiloxane 15 (hereafter "vinylmethicone/lauryldimethicone cross polymer) corresponds to INCI designation "vinyldimethicone/lauryldimethicone cross polymer". [0115]

The blend ratio of the non-emulsifying

cross-linked silicone is preferably 0.5-5.0 wt% of the total amount of the skin treatment composition. If the blend ratio is less than 0.5 wt% then the effect of the present invention is hard to obtain; on the other hand, adding more than 5.0 wt% would

not increase the effect and stickiness would

result.

[0116]

25

Selection of the liquid oil component used in the present invention is not limited in

5 particular as long as it is a liquid oil component that can be blended into a liquid cosmetic at an ordinary temperature (25°C). Examples of the liquid oil component include liquid silicone oil, liquid hydrocarbon oil, liquid ester oil, and

10 liquid higher aliphatic acid.

[0117]

Examples of said liquid silicone oil include straight chain or cyclic silicone oil; specific examples include dimethyl silicone,

15 decamethylcyclopentasiloxane,
 octamethylcyclotetrasiloxane, phenyl dimethicone,
 and octyl trimethicone.
 [0118]

Examples of said liquid ester oil include glyceryl tri-2-ethylhexanoate, diisobutyl adipate, di-2-ethylhexyl succinate, cetyl 2-ethylhexanoate,

- 2-hexyldecyl 2-ethylhexanoate, neopentyl glycol di-2-ethylhexanoate, glyceryl tri-2-ethylhexanoate, trimethylolpropane tri-2-ethylhexanoate, glyceryl tri- (caprylate/caprate), neopentyl glycol
- dicaprate, 2-ethylhexyl isononanoate, isononyl isononanoate, isodecyl isononanoate, isotridecyl isononanoate, 2-octyldecyl myristate, isopropyl palmitate, 2-ethylhexyl palmitate, 2-hexyldecyl stearate, cetyl isostearate, isopropyl isostearate,
- 10 2-hexyldecyl isostearate, isostearyl isostearate, isodecyl pivalate, isostearyl pivalate, 2-octyldodecyl pivalate, 2-octyldodecyl dimethyloctanoate, 2-ethylhexyl hydroxystearate, 2-octyldodecyl 12-stearoylstearate, oleyl oleate,
- 15 2-ethylhexyl salicylate, and jojoba oil.
  [0120]

Examples of said liquid higher aliphatic acid include isostearic acid.

[0121]

- The liquid oil component in the present invention is blended in as a constituent ingredient of the skin treatment composition of the present invention; it also functions as a swelling agent of said non-emulsifying cross-
- 25 linked silicone.

[0122]

In the present invention, when preparing the skin treatment composition, it is preferable to blend in the non-emulsifying cross-linked silicone in a swollen form (gel-like composition) swollen with said liquid oil component. This way, a skin treatment composition having a superior effect can be prepared in a stable form.

[0123]

5

For the liquid oil component for this purpose, a liquid oil component having a low viscosity at ordinary temperatures, 100 mPa·s or less for example, is preferable. A preferable viscosity range is 1-100 mPa·s.

15 [0124]

20

In the non-emulsifying cross-linked silicone swollen with the liquid oil, which is a preferable form for blending into the present invention, a preferable mass ratio of the non-emulsifying cross-linked silicone and the liquid oil is 5-40:95-60. Within this mass ratio range it is a preferable swollen material for the skin treatment composition of the present invention.

[0125]

25 Said swollen form of the non-emulsifying

cross-linked silicone is commercially available and therefore such commercial products can be used, examples of which follow.

[0126]

- Examples of a swollen form of INCI 5 designation dimethicone/vinyldimethicone cross polymer or polysilicone-11 include KSG-15 ((a mixture of dimethicone/vinyldimethicone) cross polymer and cyclopentasiloxane, approximately 5% 10 of which is cross-linked), KSG-16 ((a mixture of dimethicone/vinyldimethicone) cross polymer and dimethicone 6 mPa·s, approximately 25% of which is cross-linked), KSG-18 ((a mixture of dimethicone/vinyldimethicone) cross polymer and 15 phenyltrimethicone, approximately 15% of which is cross-linked) (these are from Shin-Etsu Chemical Co., Ltd.), GRANSIL GCM (a mixture of polysilicone-11 and octamethylcyclotetrasiloxane, approximately 6% of which is cross-linked),
- 20 GRANSIL GCM-5 (a mixture of polysilicone-11 and decamethylcyclopentasiloxane, approximately 6% of which is cross-linked), GRANSIL IDS (a mixture of polysilicone-11 and isodecane, approximately 7% of which is cross-linked), GRANSIL DMG-6 (a mixture of polysilicone-11 and dimethicone 6 mPa·s,

approximately 18% of which is cross-linked),
GRANSIL DMG-20 (a mixture of polysilicone-11 and
dimethicone 20 mPa·s, approximately 25% of which
is cross-linked), GRANSIL DMG-50 (a mixture of

5 polysilicone-11 and dimethicone 50 mPa·s,
approximately 26% of which is cross-linked), and
GRANSIL PM (a mixture of polysilicone-11 and
phenyltrimethicone, approximately 20% of which is
cross-linked), and GRANSIL ININ (a mixture of

10 polysilicone-11 and isononyl isononanoate,
approximately 15% of which is cross-linked) (these
are from GRANT Inc.).
[0127]

designation vinyldimethicone/lauryldimethicone
cross polymer include KSG-41 ((a mixture of
vinyldimethicone/lauryldimethicone) cross polymer
and liquid petrolatum, approximately 30% of which
is cross-linked), KSG-42 ((a mixture of
vinyldimethicone/lauryldimethicone) cross polymer
and light isoparaffin, approximately 25% of which
is cross-linked), KSG-43 ((a mixture of
vinyldimethicone/lauryldimethicone) cross polymer
and glyceryl tri-2-ethylhexanoate, approximately
30% of which is cross-linked), and KSG-44 ((a

Examples of a swollen form of INCI

mixture of vinyldimethicone/lauryldimethicone) cross polymer and squalane, approximately 5% of which is cross-linked), all of which are from Shin-Etsu Chemical Co., Ltd.

5 [0128]

Examples of a swollen form of INCI
designation dimethicone cross polymer include
DC9040 (a mixture of dimethicone cross polymer and
decamethylcyclopentasiloxane, approximately 12% of
which is cross-linked), DC9041 (a mixture of
dimethicone cross polymer and dimethicone 5 mPa·s,
approximately 16% of which is cross-linked), and
DC9045 (a mixture of dimethicone cross polymer and
decamethylcyclopentasiloxane, approximately 12.5%
of which is cross-linked), all of which are from
Dow Corning Toray.

[0129]

The blend ratio of the liquid oil component is preferably 0.5-30 wt% of the total amount of

the skin treatment composition. Within this blend ratio range, the effect of the present invention can be sufficiently obtained.

The skin treatment composition for wrinkle 25 reduction of the present invention further contains as an essential ingredient a water insoluble film forming polymer having a film shrinkage rate of 20% or less; the main ingredient of said film forming polymer is polyurethane having a film shrinkage rate of 20% or less. The film shrinkage rate is defined as described above. [0131]

In the present invention, the film forming polymer preferably consists of polyurethane having a film shrinkage rate of 20% or less. However, the present invention does not reject the addition of other film forming polymers within the range that does not adversely affect the effect of the present invention, provided that the film shrinkage rate of the total film forming polymer is 20% or less.

[0132]

5

Those for which said film shrinkage rate is over 20% have a poor wrinkle reduction effect,

20 peel off the skin, cause noticeable "gloss", and exhibit poor usability due to discomfort; therefore they cannot manifest the effects of the present invention. The preferable range of the film shrinkage rate is 10% or less. The lower

25 limit of the film shrinkage rate is 0, i.e. no

shrinkage, which is preferable; however, 5% is sufficient and also industrially practical. That is, the film shrinkage rate in the present invention is 0-20%; within this range, preferable combinations of the upper and lower limits are used, examples include 5-20%, 0-10%, and 5-10%.

[0133]

5

The polyurethane used in the present invention is a polymer having urethane bonds; the urethane bond is formed by an addition reaction between an isocyanate group and a compound having active hydrogen, such as a hydroxyl group. The polyurethane in the present invention is preferably obtained with a conventional method reacting at least (A) an isocyanate compound having two isocyanate groups and (B) a diol compound having two hydroxyl groups.

[0134]

Selection of said isocyanate compound

(ingredient A) is not limited in particular as long as it is used in conventional polyurethane manufacturing, examples for which include organic diisocyanate compounds such as aliphatic diisocyanate compounds, alicyclic isocyanate compounds.

More preferable are aliphatic diisocyanate compounds and alicyclic diisocyanate compounds.

One, two or more isocyanate compounds are freely selected and used.

## 5 [0135]

Examples of said aliphatic diisocyanate compounds include ethylene diisocyanate, 2,2,4-trimethylhexamethylene diisocyanate, and 1,6-hexamethylene diisocyanate.

## 10 [0136]

15

20

25

Examples of said alicyclic diisocyanate compounds include hydrogenated 4,4'-diphenylmethane diisocyanate, 1,4-cyclohexane diisocyanate, methylcyclohexylene diisocyanate, isophorone diisocyanate (hereafter referred to as "IPDI"), and norbornane diisocyanate.

Examples of said aromatic diisocyanate compound include 4,4'-diphenylmethane diisocyanate, xylylene diisocyanate, toluene diisocyanate, and naphthalene diisocyanate.

[0138]

Of the specific examples of said isocyanate compound (ingredient A), 1,6-hexymethylene diisocyanate, IPDI, and norbornane diisocyanate

are preferable due to superior weather resistance and availability. IPDI is particularly preferable.

[0139]

Selection of said diol compound (ingredient B) is not limited in particular as long as it is 5 used in conventional polyurethane manufacturing, preferable examples for which include an alkylene diol, carboxyl group-containing alkyl diol, alicyclic diol, spiro diol, polyester diol, 10 polyether diol, polycarbonate diol, polybutadiene diol, polyisoprene diol, and polyolefin diol. these, particularly preferably used are alkylene diol, carboxyl group-containing alkylene diol, alicyclic diol, polyether diol, and polycarbonate One, two or more diol compounds are freely 15 diol. selected and used.

[0140]

20

25

Examples of said alkylene diol include ethylene glycol, propylene glycol, 1,4-butane diol, 1,6-hexane diol, neopentyl glycol, 1,8-octane diol, and 1,10-decane diol.

[0141]

Preferable examples of said carboxyl groupcontaining alkylene diol include carboxylic acid having 3-26 carbons, more preferably 3-12 carbons, as well as having a dialylol groups, such as dimethylol, diethanol and propanol. Specific examples include dimethylol propionic acid (hereafter "DMPA") and dimethylol butanoic acid (hereafter "DMBA"); a mixture thereof can be used as well.

[0142]

5

In the present invention, when a carboxyl group-containing alkylene diol is used for the

10 diol compound in the synthesis to obtain polyurethane containing carboxyl groups in the molecule, the carboxyl groups incorporated in the molecule can be neutralized by neutralizers such as triethylamine, trimethylamine, 2-amino-2
15 methyl-1-propanol, triethanolamine, potassium hydroxide, and sodium hydroxide to obtain a stable water dispersion containing polyurethane, which is a preferred embodiment described later.

[0143]

Examples of said alicyclic diol include 1,4-cyclohexane dimethanol (hereafter "CHDM"), which is preferable because it gives adequate strength to the film.

[0144]

25 Preferable examples of said spiro diol

include spiro glycol.
[0145]

Examples of said polyester diol include those obtained by condensation polymerization between al least one chosen from a group of 5 dicarboxylic acids including succinic acid, glutaric acid, adipic acid, sebacic acid, azelaic acid, maleic acid, fumaric acid, phthalic acid, and terephthalic acid and at least one chosen from a group of diols including ethylene glycol, 10 propylene glycol, 1,4-butane diol, 1,6-hexane diol, neopentyl glycol, 1,8-octane diol, 1,10-decane diol, diethylene glycol, polyethylene glycol (hereafter "PEG"), polypropylene glycol (hereafter "PPG"), tetramethylene glycol, polytetramethylene 15 glycol, and spiro glycol, as well as those obtained by the ring opening polymerization of lactonic acid.

[0146]

Examples of said polyether diol include

polyether diols contained in diols used in the

synthesis of said polyester diol, examples of

which include polyoxyethylene diols such as

diethylene glycol and PEG, polyoxypropylene diols

such as PPG, polyoxytetramethylene diols such as

polytetramethylene glycol (hereafter "PTMG"),
phenols such as bisphenol A, and a product of the
ring opening addition polymerization of bisphenol
A and at least one compound chosen from propylene
oxide (hereafter "PO") and ethylene oxide
(hereafter "EO") (if a copolymer is used, such a
copolymer can be either a block copolymer or
random copolymer). Of these, PEG, PPG, and PTMG
are preferable, and PTMG is particularly
preferable since it forms a soft film and adheres
to the skin very well.
[0147]

Preferable examples of said polycarbonate diol include polyhexamethylene carbonate diol (hereafter "PHMC") because of its ability to form a soft film and its superior adhesion to the skin.

15

In the present invention, polyurethane that forms a film that manifests a superior wrinkle

20 reduction effect can be obtained by using the diol compound (ingredient B) consisting of a mixture of a polyether diol, polycarbonate diol, and carboxyl group-containing alkylene diol.

[0149]

In the present invention, when mixing a

polyether diol, polycarbonate diol, and carboxyl group-containing alkylene diol, it is particularly preferable to use PTMG for said polyether diol, PHMC for the polycarbonate diol, and DMPA and/or DMBA for said carboxyl group-containing alkylene diol.

[0150]

5

In the present invention, when using the aforementioned preferable diol compounds, it is

10 particularly preferable to use an isocyanate compound that contains IPDI; a skin treatment composition for wrinkle reduction having the most exceptional wrinkle reduction effect can be obtained by using polyurethane synthesized by

15 using IPDI for the isocyanate and PTMG, PHMC, DMPA and/or DMBA for the diol compounds.

The molar ratio of said isocyanate compound (ingredient A) and diol compound (ingredient B) is preferably A/B = 2/0.8-2/1.8, and more preferably A/B = 2/1-2/1.8.

[0152]

20

25

In the present invention, it is preferable to use polyurethane having structural units derived from alkylene oxide (hereafter "RO"); this

makes it easier to control the elongation of the film obtained from the water dispersion of the polyurethane that is a preferred embodiment described later to obtain a flexible film. A skin treatment composition that manifests superior effects and superior usability can be obtained by using this polyurethane for the preparation.

[0153]

5

Examples of the compound having a structural 10 unit derived from RO include a polyoxyethylene diol such as diethylene glycol and PEG, a polyoxypropylene diol such as PPG, a polyoxytetramethylene diol such as PTMG, polyoxyethylene polyoxypropylene glycol (EO/PO block copolymer), phenols such as bisphenol A, and 15 a product of the ring opening addition polymerization of bisphenol A and at least one compound chosen from PO and EO (if a copolymer is used, such a copolymer can be either a block 20 copolymer or random copolymer); preferably used are PEG, PPG, PTMG, etc. [0154]

In the present invention, the compound.

having a structural unit derived from RO is used

25 as a polyether diol ingredient in said ingredient

Β.

[0155]

is preferably blended in as a water dispersion,
i.e. dispersed in water, which is an essential

ingredient of the present invention. Said water
dispersion, when applied on the skin and dried,
forms a polymer film on the skin.

[0156]

In the present invention, when preparing the

10 skin treatment composition, the film forming

polymer

[0157]

The water dispersion of polyurethane is prepared with a conventional method; for example,

15 a pre-polymer having remaining isocyanate obtained from a reaction in an organic solvent is dispersed in water containing potassium hydroxide under high speed agitation, followed by a chain elongation reaction to increase the molecular weight, and

20 said organic solvent is recovered from the obtained water based solution to obtain the water dispersion of the polyurethane.

[0158]

In the present invention, the following is
25 particularly preferable for said polyurethane and

water dispersion of polyurethane. That is: [0159]

the polyurethane, the strength is 300-700 kg/cm²,

and preferably 400-600 kg/cm². If the strength is less than 300 kg/cm², then the film cannot follow the skin movements and peeling easily occurs. If it is over 700 kg/cm², then the film causes discomfort on the skin. The elongation is 200
500%, preferably 300-500%. If the elongation is less than 200%, then the film formed on the skin cannot follow the skin movements very well. If it is over 500%, then the wrinkle reduction effect becomes insufficient.

# 15 [0160]

The average particle size of the polyurethane in the water dispersion of the polyurethane is 10-300 nm, preferably 20-200 nm.

If the average particle size is less than 10 nm,

then there is a sufficient small-wrinkle reduction effect but on the other hand the large-wrinkle reduction effect becomes insufficient. If the average particle size is over 300 nm, then adhesion to the skin becomes poor and peeling

[0161]

A preferable embodiment in the present invention uses two kinds of water dispersions of polyurethane having different particle sizes.

5 This way, not only the wrinkle reduction effect but also a skin mark erasing effect can be obtained. For said two kinds of polyurethane having different particle sizes, polyurethanes having an average particle size of 20-60 nm and an average particle size of 150-200 nm are preferable.

[0162]

Preparation examples of the polyurethane having a film shrinkage rate of 20% or less used in the present invention are described below.

**15** [0163]

(Preparation example 2-1)

weight 1,000), 5 g of CHDM, and 10 g of DMBA were put into a four-neck flask equipped with a stirrer, a thermometer, a nitrogen introduction tube, and a reflux cooling apparatus, to which 50 g of ethyl acetate was added as a solvent, and then an oil bath was used to raise the temperature to 80℃ and allow the reaction to proceed for 6 hours to

groups. This pre-polymer having remaining isocyanate groups was cooled down to 50°C and dispersed in 800 g of water containing 6 g of potassium hydroxide under high speed stirring,

- 5 followed by 3 hours of a chain elongation reaction at 50℃ to increase the molecular weight. Said ethyl acetate was recovered from the obtained water-based liquid to obtain a water dispersion of polyurethane substantially having no solvent
- 10 (polyurethane solid content 20 wt%).
   (Average particle size: 170 nm, film strength: 410
   kg/cm², film elongation: 320%, film shrinkage
   rate: 11%)
   [0164]

15 (Preparation example 2-2)

50 g of IPDI, 60 g of PTMG (molecular weight 1,000), 40 g of PHMC (molecular weight 2,000), and 10 g of DMBA were put into a four-neck flask equipped with a stirrer, a thermometer, a nitrogen introduction tube, and a reflux cooling apparatus, to which 50 g of ethyl acetate was added as a solvent, and then an oil bath was used to raise the temperature to 80℃ and allow the reaction to proceed for 3 hours. 2 g of N-

25 methyldiethanolamine (NMDE tA) and 40 g of ethyl

acetate were added, followed by 3 hours of reaction at 80°C to obtain a pre-polymer having remaining isocyanate groups. This pre-polymer having remaining isocyanate groups was cooled down to 50°C and dispersed in 700 g of water containing 6 g of potassium hydroxide under high speed stirring, followed by 3 hours of a chain elongation reaction at 50°C to increase the molecular weight. Said ethyl acetate was

10 recovered from the obtained water-based liquid to obtain a water dispersion of polyurethane substantially having no solvent (polyurethane solid content 20 wt%).

(Average particle size: 40 nm, film strength: 530
15 kg/cm², film elongation: 360%, film shrinkage
 rate: 9%)

[0165]

The blend ratio of the polyurethane having a film shrinkage rate of 20% or less in the present invention is preferably 0.1-10.0 wt% of the total amount of the skin treatment composition. If the blend ratio is less than 0.1 wt% then the effect of the present invention is hard to be obtained sufficiently; and if it is over 10.0 wt% then the film tends to peel off the skin. A more

preferable blend ratio range is 1.0-8.0 wt% of the total amount of the skin treatment composition.

[0166]

As mentioned before, the skin treatment

5 composition of the present invention can contain, in addition to the polyurethane having a film shrinkage rate of 20% or less, other film forming polymers, examples of which include acrylic type polymers. Even when film forming polymers other

10 than the polyurethane having a film shrinkage rate of 20% or less are added, the blend ratio of the total film forming polymers is still preferably in the range of 0.1-10.0 wt%.

[0167]

The other essential ingredient of the present invention is water. Water other than the water contained in said essential ingredients is further added as appropriate to form the skin treatment composition.

### 20 [0168]

25

In addition to the aforementioned ingredients, other ingredients used in skin treatment compositions such as cosmetics and drugs can be blended as necessary into the skin treatment composition of the present invention as

long as the effect of the present invention is not adversely affected. The aforementioned optional ingredients include, albeit partially redundant, oil components, power ingredients, surfactants,

humectants, water soluble polymers, thickeners, ultraviolet absorbents, sequestering agents, sugars, amino acids, organic amines, pH adjustment agents, nutritional supplements for skin, vitamins, antioxidants, and perfumes that are not those

[0169]

[0170]

mentioned above.

10

15

The skin treatment composition for wrinkle reduction of the present invention can be prepared by blending in the aforementioned ingredients and following a conventional method.

The skin treatment composition of the present invention is used in foundation cosmetic formulations in the form of a cream, emulsion,

20 lotion, or gel.

EXAMPLES

[0171]

[Invention of claims 1-12]

25 (Preparation example 1-1)

Synthesis of a water dispersion of polyurethane

50 g of IPDI, 120 g of PTMG (molecular weight 1,000), 5 g of CHDM, and 10 g of DMBA were put into a four-neck flask equipped with a stirrer,

- 5 a thermometer, a nitrogen introduction tube, and a reflux cooling apparatus, to which 50 g of ethyl acetate was added as a solvent, and then an oil bath was used to raise the temperature to 80℃ and allow the reaction to proceed for 6 hours to
- obtain a pre-polymer having remaining isocyanate groups. This pre-polymer having remaining isocyanate groups was cooled down to 50°C and dispersed in 800 g of water containing 6 g of potassium hydroxide under high speed stirring,
- 15 followed by 3 hours of a chain elongation reaction at 50℃ to increase the molecular weight. Said ethyl acetate was recovered from the obtained water-based liquid to obtain a water dispersion of polyurethane substantially having no solvent
- 20 (polyurethane solid content 20 wt%).
   (Average particle size: 170 nm, film strength: 410
   kg/cm², film elongation: 320%, film shrinkage
   rate: 11%)
  [0172]
- 25 (Preparation example 1-2)

Synthesis of a water dispersion of polyurethane 50 g of IPDI, 60 g of PTMG (molecular weight 1,000), 40 g of PHMC (molecular weight 2,000), and 10 g of DMBA were put into a four-neck flask equipped with a stirrer, a thermometer, a nitrogen 5 introduction tube, and a reflux cooling apparatus, to which 50 g of ethyl acetate was added as a solvent, and then an oil bath was used to raise the temperature to 80% and allow the reaction to 10 proceed for 3 hours. 2 g of Nmethyldiethanolamine (NMDE tA) and 40 g of ethyl acetate were added, followed by 3 hours of reaction at 80℃ to obtain a pre-polymer having remaining isocyanate groups. This pre-polymer 15 having remaining isocyanate groups was cooled down to 50°C and dispersed in 700 g of water containing 6 g of potassium hydroxide under high speed stirring, followed by 3 hours of a chain elongation reaction at  $50^{\circ}$ C to increase the 20 molecular weight. Said ethyl acetate was recovered from the obtained water-based liquid to

- recovered from the obtained water-based liquid to obtain a water dispersion of polyurethane substantially having no solvent (polyurethane solid content 20 wt%).
- 25 (Average particle size: 40 nm, film strength: 530

kg/cm<sup>2</sup>, film elongation: 360%, film shrinkage rate: 9%)

[0173]

(Preparation example 1-3)

- 5 Synthesis of a water dispersion of an acrylic type polymer (acrylic type polymer emulsion)
  100 g of ion-exchanged water and 2 g of polyoxyethylenecetyl ether were put into a fourneck flask equipped with stirring blades, a
- thermometer, a nitrogen introduction tube, and a reflux cooling apparatus; the mixture was heated and stirred as nitrogen gas was instilled into it and the liquid temperature was maintained at 80°C.

  Meanwhile, a monomer mixed solution consisting of
- 15 40 g of ion-exchanged water, 1 g of sodium
  laurylsulfate, 1 g of polyoxyethylenecetyl ether,
  50 g of methyl methacrylate, 42 g of 2-ethylhexyl
  acrylate, 3 g of methacrylic acid, and 1.4 g of
  sulfonated polyvinyl alcohol and an initiator
- aqueous solution containing 0.3 parts potassium persulfate and 10 parts ion-exchanged water were prepared. 5 wt% of the monomer mixed solution and 10 wt% of the initiator aqueous solution were added to a four-neck flask and stirred to initiate
- 25 an emulsification polymerization reaction; the

rest of the monomer mixed solution and the initiator aqueous solution were concurrently dripped into the four-neck flask over a period of approximately three hours. Stirring was continued

- for another hour as the liquid temperature was kept at 80% and then the obtained reaction mixture was cooled down to 50%. Aqueous ammonia was then added to adjust the pH to approximately 8 and the temperature was cooled down to room
- 10 temperature to obtain the target water dispersion of acrylic type polymer (acrylic type polymer solid content 50 wt%).

(Average particle size: 480 nm, film strength: 20  $\,$  kg/cm², film elongation: 700%, film shrinkage

15 rate: 6%)

[0174]

(Preparation example 1-4)

Synthesis of a water dispersion of an acrylic type polymer (acrylic type polymer emulsion)

20 100 g of ion-exchanged water and 2 g of polyoxyethylenestearyl ether were put into a fourneck flask equipped with stirring blades, a thermometer, a nitrogen introduction tube, and a reflux cooling apparatus; the mixture was heated 25 and stirred as nitrogen gas was instilled into it

and the liquid temperature was maintained at  $80\,\%$  . Meanwhile, a monomer mixed solution consisting of 40 g of ion-exchanged water, 1 g of sodium polyoxyethylene laurylsulfate, 1 g of polyoxyethylene stearyl ether, 30 g of methyl methacrylate, 67 g of 2-ethylhexyl acrylate, and 3 g of methacrylic acid and an aqueous solution containing 0.3 parts potassium persulfate and 10 parts ion-exchanged water were prepared. 5 wt% of the monomer mixed solution and 10 wt% of the initiator aqueous solution were added to a fourneck flask and stirred to initiate an emulsification polymerization reaction; the rest of the monomer mixed solution and the initiator aqueous solution were concurrently dripped into the four-neck flask over a period of approximately three hours. Stirring was continued for another hour as the liquid temperature was kept at  $80^{\circ}$ C and then the obtained reaction mixture was cooled down to 50°C. Aqueous ammonia was then added to adjust the pH to approximately 8 and the temperature was cooled down to room temperature to obtain the target water dispersion of an acrylic

5

10

15

20

25

50 wt%).

69

type polymer (acrylic type polymer solid content

(Average particle size: 100 nm, film strength: 50 kg/cm², film elongation: 500%, film shrinkage rate: 7%)

[0175]

- 5 (Method for evaluating/measuring the film)
  - 1. Film shrinkage rate

A water dispersion of a solid equivalent 1 g of the polymer (polyurethane, acrylic type polymer, etc.) was poured into a 5 cm x 5 cm polyethylene

10 mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature (50°C) for a prescribed amount of time (three days at room temperature) to obtain a film, and vertical height and horizontal length of this film

15 were measured and used in the following formula to calculate the film shrinkage rate.

[0176]

Film shrinkage rate (%) = [(Vertical measurement x Horizontal measurement)/25] x 100

20 [0177]

25

2. Measurement of the strength and elongation of the film

A water dispersion of a solid equivalent 1 g of the polymer (polyurethane, acrylic type polymer, etc.) was poured into a 5 cm x 5 cm polyethylene

mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature  $(50\,^\circ\text{C})$  for a prescribed amount of time (three days at room temperature) to obtain a film, and this

film was cut out using a dumbbell #3 for the measurement. The autograph function of "Tensile tester RTM-250" from Orientec Co., Ltd. was used to carry out the measurement of the strength and elongation at 20°C and a cross head speed of 300

10 mm/min.

[0178]

3. Particle size measurement of the water dispersion

The water dispersion of the polymer

15 (polyurethane, acrylic type polymer, etc.) was measured with a laser light scattering particle size distribution measuring apparatus from Otsuka Electronics Co., Ltd.

[0179]

The present invention is described in detail below by referring to Examples. The blend ratios are in wt% units. Before the description of Examples, the efficacy test method used in the present invention is described.

25 [0180]

(Sensory test)

Each specimen was tested by a panel of 10 specialists for the sensation during use. Each specimen was evaluated based on the following

5 criteria for usability and the sensation during use, including (1) the wrinkle reduction effect right after application, (2) the wrinkle reduction effect five hours after application, (3) peeling off the skin (right after application, three hours after application), (4) glossiness (skin greasy and shining), (5) no stickiness (right after application, three hours after application), and (6) no discomfort.

[0181]

- 15 "Evaluation criteria"
  - (1) Wrinkle reduction effect right after application
  - © : 8 or more felt a wrinkle reduction effect.
  - O: 5-7 felt a wrinkle reduction effect.
- 20  $\triangle$ : 3-4 felt a wrinkle reduction effect.
  - imes : 2 or fewer felt a wrinkle reduction effect. [0182]
  - (2) Wrinkle reduction effect five hours after application
- 25 ©: 8 or more felt a wrinkle reduction effect.

- O: 5-7 felt a wrinkle reduction effect.
- $\triangle$  : 3-4 felt a wrinkle reduction effect.
- X : 2 or fewer felt a wrinkle reduction effect.

[0183]

- 5 (3) Peeling off from the skin
  - ◎ : 8 or more felt there was no peeling off.
  - $\bigcirc$  : 5-7 felt there was no peeling off.
  - $\triangle$ : 3-4 felt there was no peeling off.
  - X : 2 or fewer felt there was no peeling off.
- 10 [0184]
  - (4) Glossiness
  - : 8 or more felt there was no glossiness.
  - $\bigcirc$  : 5-7 felt there was no glossiness.
  - $\triangle$  : 3-4 felt there was no glossiness.
- 15 imes : 2 or fewer felt there was no glossiness.

[0185]

- (5) No stickiness
- : 8 or more felt there was no stickiness.
- O: 5-7 felt there was no stickiness.
- 20  $\triangle$ : 3-4 felt there was no stickiness.
  - × : 2 or fewer felt there was no stickiness.

[0186]

- (6) No discomfort
- : 8 or more felt there was no discomfort.
- 25  $\bigcirc$ : 5-7 felt there was no discomfort.

 $\triangle$  : 3-4 felt there was no discomfort.

imes : 2 or fewer felt there was no discomfort. [0187]

"Examples 1-1 to 1-13, Comparative examples 1-1 to 1-9"

Skin treatment compositions for wrinkle reduction were prepared by mixing ingredients with blend ratios (total blend ratio 100 wt%) according to the recipes shown in Tables 1-1 to 1-3. Also, the efficacy test was conducted on the skin treatment compositions for wrinkle reduction from the aforementioned Examples 1-1 to 1-13 and Comparative examples 1-1 to 1-9; the evaluation results are also shown in Tables 1-1 to 1-3.

[0188]

# [Table 1-1]

				E	хатрІ	e			
	1-1	1-2	1-3	1-4	1-5	1-6	1-7	1-8	1-9
Ion-exchanged water	Balance								
Glycerin	6.0	6. 0	6. 0	6. 0	6. 0	6. 0	6. 0	6.0	6. 0
Water dispersion of polyurethane (solid content 20	10.0	5. 0	10. 0	-	-	5. 0	45. 0	10. 0	10. 0
wt%) (preparation example 1)									
Water dispersion of	_	5.0	-	10.0	10. 0	_	_	10. 0	20. 0
polyurethane (solid content 20 wt%) (preparation example 2)									
Water dispersion of an acrylic type polymer (solid content 50 wt%) (preparation example 3)	10. 0	10. 0	_	10.0	_	5. 0	-	15. 0	10.0
Water dispersion of an acrylic type polymer (solid content 50 wt%) (preparation example 4)	_	-	10. 0	-	10. 0	-	30. 0	_	10.0
Total	100	100	100	100	100	100	100	100	100
Wrinkle reduction effect right after application	0	0	0	0	0	0	0	0	0
Wrinkle reduction effect five hours after application	0	0	0	0	0	0	0	0	0
Peeling	0	0	0	0	0	0	0	0	0
Glossiness	0	0	0	0	0	0	0	0	0
No stickiness	0	0	0	0	0	0	0	0	0
No discomfort	0	0	0	0	0	0	0	0	0

[0189]

# [Table 1-2]

		Exar	прІе	
	1-10	1-11	1-12	1-13
Ion-exchanged water	Balance	Balance	Balance	Balance
Glycerin	6. 0	6. 0	6. 0	6.0
Water dispersion of	5.0	5.0	5. 0	5.0
polyurethane (solid content 20				
wt%)(preparation example 1)				
Water dispersion of	5.0	5.0	5.0	5.0
polyurethane (solid content 20				
wt%)(preparation example 2)				
Water dispersion of an acrylic	10.0	5. 0	10.0	5.0
type polymer (solid content 50				
wt%) (preparation example 3)				
Water dispersion of an acrylic	-	5.0	-	5.0
type polymer (solid content 50		1		
wt%) (preparation example 4)				
Water dispersion of scaly	10.0	10.0	4. 0	20. 0
silica (solid content 15 wt%)				
(Note 6)				
Total	100	100	100	100
Wrinkle reduction effect right	0	0	0	0
after application				
Wrinkle reduction effect five	0	0	0	0
hours after application				
Peeling	0	0	0	0
Glossiness	0	0	<b>O</b> .	0
No stickiness	0	0	0	0
No discomfort	0	0	0	0

[0190]

[Table 1-3]

	Comparative example								
	1-1	1-2	1-3	1-4	1-5	1-6	1-7	1-8	1-9
Ion-exchanged water	Balance	Balance	Balance	Balance	Balance	Balance	Balance	Balance	Balance
Glycerin	6.0	6.0	6.0	6.0	6. 0	6.0	6. 0	6. 0	6. 0
Water dispersion of	-	_	-	10.0	10. 0	-	-	-	_
polyurethane (solid content 20									
wt%)(preparation example 1)									
Water dispersion of an acrylic	-		-	_	_	-	-	10.0	10.0
type polymer (solid content 50									
wt%) (preparation example 3)									
Commercially available water	10.0	-	-	_	_	-	-	-	-
dispersion of polyurethane									
(solid content 33 wt%) (Note 1)									
Commercially available water	-	10.0	-	_	-	-	-	_	-
dispersion of polyurethane			ļ					ļ	
(solid content 39 wt%) (Note 2)									
Commercially available water	_	-	10.0	-	_	-	-	-	10. 0
dispersion of polyurethane									
(solid content 20 wt%) (Note 3)									
Commercially available water	_	-	-	-	10. 0	10. 0	-	-	_
dispersion of an acrylic type	1								
polymer (solid content 50 wt%)									
(Note 4)									
Commercially available water	_	_	-	-	-	-	10.0	-	-
dispersion of an acrylic type									
polymer (solid content 48 wt%)					ĺ				
(Note 5)									
Total	100	100	100	100	100	100	100	100	100
Wrinkle reduction effect right	0	0	0	Δ	0	0	0	Δ	0
after application									
Wrinkle reduction effect five	×	×	×	×	×	×	×	×	×
hours after application								ļ	
Peeling	×	×	×	Δ	×	×	×	Δ	×
Glossiness	×	×	×	Δ	×	×	×	Δ	×
No stickiness	0	0	0	0	×	×	×	×	×
No discomfort	×	×	×	0	×	×	×	0	×

[0191]

In Tables 1-1 to 1-3:

- (Note 1) Avalure UR405 (from NOVEON)
- (Average particle size: 100 nm, strength: 40  $\,$  kg/cm², elongation: 150%, film shrinkage rate: 35%)
- 5 (Note 2) Avalure UR445 (from NOVEON)
  - (Average particle size: 5 nm, strength: 240  $kg/cm^2$ , elongation: 610%, film shrinkage rate: 27%)
  - (Note 3) Iodosol PUD (from Nippon NSC)
- 10 (Average particle size: 20 nm, strength: 200 kg/cm², elongation: 530%, film shrinkage rate: 23%)
  - (Note 4) Dashcoat (from Daito Kasei Kogyo Co., Ltd.)
- (Average particle size: 150 nm, strength: 15  $kg/cm^2$ , elongation: 700%, film shrinkage rate: 25%)
  - (Note 5) Ultrasol 2075C (from Ganz Chemical Co., Ltd.)
- 20 (Average particle size: 200 nm, strength: 40  $kg/cm^2$ , elongation: 1000%, film shrinkage rate: 13%)
  - (Note 6) Sunlovely LFS-C (solid content 15%) (from Dohkai Chemical Industries Co., Ltd.)
- **25** [0192]

As clearly shown in Tables 1-1 to 1-3, the skin treatment compositions for wrinkle reduction from Examples 1-13 pertaining to the present invention are superior in terms of the wrinkle reduction effect and free of peeling, glossiness, stickiness, and discomfort. On the contrary, skin treatment compositions for wrinkle reduction from Comparative examples 1-9, which do not meet the constituent requirements of the present invention, do not manifest the effects of the present invention.

[0193]

5

10

15

20

More Examples of the skin treatment composition for wrinkle reduction of the present invention are described below. The aforementioned efficacy test was conducted on these and the results were superior for all of these.

[0194]

[Example 1-14] Skin treatment composition for wrinkle reduction (cream type)

		Ingredients	Blend	ratio (wt%)
	(1)	Stearyl alcohol		6.0
	(2)	Stearic acid		2.0
	(3)	Hydrated lanolin		4.0
25	(4)	Squalane		9.0

```
(5) 5-Octyldodecanol
                                               10.0
                                                6.0
    (6) 1,3-butylene glycol
    (7) Water dispersion of polyurethane from
                                                5.0
    preparation example 1
    (8) Water dispersion of polyurethane from
5
                                               10.0
    preparation example 2
    (9) Water dispersion of an acrylic type polymer
                                                8.0
    from preparation example 3
    (10) PEG 1500
                                                4.0
10
    (11) Polyoxyethylene (25) cetyl alcohol ether
                                                3.0
                                                2.0
    (12) Glycerin monostearate
                                                0.1
    (13) Ethylparaben
    (14) Butylparaben
                                                0.1
                                                0.01
15
    (15) Tocopherol
                                                0.1
    (16) Perfume
    (17) Purified water
                                               Balance
                                                5.0
    (18) Scaly silica slurry (Note 1)
                                          Total 100.0
20
    [0195]
    (Note 1) Sunlovely LFS-C (solid content 15%) (from
    Dohkai Chemical Industries Co., Ltd.)
    [0196]
    <Preparation method>
         (6), (10), (13), and (14) were added to (17)
25
```

and the temperature was raised and adjusted to 70%. Next, the oil phase consisting of (1), (2), (3), (4), (5), (11), (12), (15), and (16) were prepared and the temperature was adjusted to 70%.

5 This is added to the water phase previously prepared; a homomixer was used to homogenize the emulsified particles and (7), (8), (9), and (18) were added. After deaeration, filtration, and cooling, the target cream for wrinkle reduction was obtained.

[0197]

25

[Example 1-15] Skin treatment composition for wrinkle reduction (lotion type)

		Ingredients	Blend	ratio	(wt%)
15	(1)	Stearic acid			2.0
	(2)	Cetyl alcohol			1.5
	(3)	Petrolatum			4.0
	(4)	Squalane			5.0
	(5)	Glycerol tri-2-ethylhexanoa	ate		2.0
20	(6)	Sorbitan monooleate			2.0
	(7)	Dipropylene glycol			5.0
	(8)	PEG 1500			3.0
	(9)	Water dispersion of polyure	ethane	from	
	prep	paration example 1			2.0

(10) Water dispersion of polyurethane from

preparation example 2

8.0

1.0

0.1

- (11) Water dispersion of an acrylic type polymer from preparation example 4 10.0
- (12) Triethanolamine
- 5 (13) Methylparaben
  - (14) Phenoxy ethanol 0.1
  - (15) Perfume 0.1
  - (16) Purified water Balance

Total 100.0

#### 10 [0198]

<Preparation method>

- (7), (8), (12), (13), and (14) were added to (16) and the temperature was raised and adjusted to 70%. The oil phase consisting of (1), (2),
- 15 (3), (4), (5), (6), and (15) were prepared and the temperature was adjusted to 70°C. This oil phase was added to the water phase previously prepared and pre-emulsification was carried out. After homogenizing the emulsified particles with a
- 20 homomixer, (9), (10), and (11) were added to obtain the target lotion for wrinkle reduction.

  [0199]

[Example 1-16] Skin treatment composition for wrinkle reduction (gel type)

25 Ingredients Blend ratio (wt%)

- 7.0 (1) Dipropylene glycol 8.0 (2) PEG 1500 0.4 (3) Carboxyvinyl polymer 0.2 (4) Methylcellulose (5) Polyoxyethylene (15) oleyl alcohol ether 5 1.0 0.1 (6) Potassium hydroxide (7) Water dispersion of polyurethane from 3.0 preparation example 1 10 (8) Water dispersion of polyurethane from 4.0 preparation example 2 (9) Water dispersion of an acrylic type polymer 5.0 from preparation example 3 (10) Edetate 0.01 0.1 15 (11) Perfume Balance (12) Purified water 0.2 (13) Ethylparaben Total 100.0 [0200]
- 20 <Preparation method>

25

(3) and (4) were homogeneously dissolved in (12), to which (2) and (10) were added. (5) was added to (1) and heated and dissolved at 55%, to which (11) and (13) were added. This was slowly added to the water phase prepared previously as

the latter was being stirred. Next, (7), (8), and

- (9) were added. Finally, an aqueous solution of
- (6) was added and thorough stirring was done for neutralization to obtain the target gel for
- 5 wrinkle reduction.

### [0201]

[Example 1-17] Skin treatment composition for wrinkle reduction (lotion type)

	W 1 1.	nkie leduction (lotion type,	,		
		Ingredients	Blend	ratio (	w t %)
10	(1)	Stearic acid		2.	0
	(2)	Cetyl alcohol		1.	5
	(3)	Petrolatum		4.	0
	(4)	Squalane		5.	0
	(5)	Glycerol tri-2-ethylhexanoa	ate	2.	0
1 5	(6)	Sorbitan monooleate		2.	0
	(7)	Dipropylene glycol		5.	0
	(8)	PEG 1500		3.	0
	(9)	Water dispersion of polyure	ethane	from	
	pre	paration example 1		0.	5
20	(10)	) Water dispersion of polyu	rethan	e from	
	pre	paration example 2		9.	5
	(11)	) Water dispersion of an act	rylic	type pol	ymer
	froi	n preparation example 3		10.	0
	(12)	) Water dispersion of cross	linke	d sodium	poly-
25	γ - 1	glutamate (Note 1)		10.	0

(13) Triethanolamine

1.0

(14) Methylparaben

0.1

(15) Phenoxy ethanol

0.1

(16) Perfume

0.1

5 (17) Purified water

Balance

Total 100.0

[0202]

(Note 1) Gelprotein A-8001 (solid content 2 wt%) (from Idemitsu Technofine Co., Ltd.)

10 [0203]

<Preparation method>

(7), (8), (12), (13), (14), and (15) were added to (17) and the temperature was raised and adjusted to 70°C. Next, the oil phase consisting of (1), (2), (3), (4), (5), (6), and (16) were prepared and the temperature was adjusted to 70°C. This oil phase was added to the water phase prepared as above and pre-emulsification was carried out. After the emulsified particles were homogenized with a homomixer, (9), (10), and (11) were added to obtain the target lotion type for wrinkle reduction.

[0204]

[Example 1-18] Skin treatment composition for wrinkle reduction (gel type)

	Ingredients	Blend rat	io (wt%)
	(1) Dipropylene gly	col	5.0
	(2) Glycerin		2.0
	(3) PEG 1500		8.0
5	(4) Carboxyvinyl po	olymer	0.4
	(5) Methylcellulose	•	0.2
	(6) Polyoxyethylene	e (15) oleyl alcohol e	ther
			1.0
	(7) Potassium hydro	oxide	0.1
10	(8) Water dispersion	on of polyurethane fro	m
	preparation example	2 1	1.0
	(9) Water dispersion	on of polyurethane fro	m
	preparation example	2	6.0
	(10) Water dispersi	on of an acrylic type	polymer
15	from preparation ex	cample 3	5.0
	(11) Water dispersi	on of cross linked so	dium poly-
	γ -glutamate (Note	1)	50.0
	(12) Edetate		0.01
	(13) Perfume		0.1
20	(14) Purified water	•	Balance
	(15) Ethylparaben		0.2
		Tota	1 100.0
	[0205]		
	(Note 1) Gelproteir	A-8001 (solid conten	t 2 wt%)
25	(from Idemitsu Tech	nnofine Co., Ltd.)	

[0206]

<Preparation method>

- (4) and (5) were homogeneously dissolved in (14) and then (2), (3), (11), and (12) were added.
- 5 (6) was added to (1) and heated and dissolved at 60°C, to which (13) and (15) were added. This was slowly added to the water phase prepared previously as the latter was being stirred. Next, (7), (8), and (10) were added. Finally, an
- 10 aqueous solution of (7) was added and thorough stirring was done for neutralization to obtain the target gel for wrinkle reduction.

[0207]

[Invention of claims 13-25]

- The present invention is described in detail below by referring to Examples. The blend ratios are in wt% units. Before the description of Examples, the method for evaluating/measuring the film and the efficacy test method used in the
- 20 present invention are described.

[0208]

[Method for evaluating/measuring the film)

(1) Film shrinkage rate

A water dispersion of a solid equivalent 1 g of the polymer is poured into a 5 cm x 5 cm

polyethylene mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature ( $50^{\circ}\text{C}$ ) for a prescribed amount of time (three days at room temperature) to obtain a film, and vertical height and horizontal length of this film are measured and used in the following formula to calculate the film shrinkage rate. [0209]

Film shrinkage rate (%) = [(Vertical

10 measurement x Horizontal measurement)/25] x 100

[0210]

5

(2) Measurement of the strength and elongation of the film

A water dispersion of a solid equivalent 1 g 15 of the polymer was poured into a 5 cm x 5 cm polyethylene mold such that the film thickness is approximately 0.5 mm and dried at a prescribed temperature (50°C) for a prescribed amount of time (three days at room temperature) to obtain a film, and this film was cut out using a dumbbell #3 for 20 The autograph function of the measurement. "Tensile tester RTM-250" from Orientec Co., Ltd. was used to carry out the measurement of the speed of 300 mm/min. 25

[0211]

(3) Particle size measurement of the water dispersion

The water dispersion of the polymer was

5 measured with a laser light scattering particle
size distribution measuring apparatus from Otsuka
Electronics Co., Ltd.

[0212]

[Sensory test]

- 10 Each specimen was used by a panel of 10 The specialists assessed each specialists. specimen for usability and the sensation during use, including (1) the wrinkle reduction effect right after application, (2) the wrinkle reduction effect five hours after application, (3) peeling 15 off the skin (right after application, three hours after application), (4) glossiness (skin greasy and shining), (5) no stickiness (right after application, three hours after application), and 20 (6) no discomfort; the evaluation was conducted by using the following criteria.
  - [0213]

"Evaluation criteria"...

- (1) Wrinkle reduction effect right after
- 25 application

- ② : 8 or more felt a wrinkle reduction effect.
- O: 5-7 felt a wrinkle reduction effect.
- $\triangle$  : 3-4 felt a wrinkle reduction effect.
- X : 2 or fewer felt a wrinkle reduction effect.
- 5 [0214]
  - (2) Wrinkle reduction effect five hours after application
  - ◎ : 8 or more felt a wrinkle reduction effect.
  - O: 5-7 felt a wrinkle reduction effect.
- 10  $\triangle$ : 3-4 felt a wrinkle reduction effect.
  - imes : 2 or fewer felt a wrinkle reduction effect.

[0215]

- (3) Peeling off from the skin
- : 8 or more felt there was no peeling off.
- 15  $\bigcirc$ : 5-7 felt there was no peeling off.
  - $\triangle$  : 3-4 felt there was no peeling off.
  - $\times$  : 2 or fewer felt there was no peeling off.

[0216]

- (4) Glossiness
- 20 © : 8 or more felt there was no glossiness.
  - $\bigcirc$  : 5-7 felt there was no glossiness.
  - $\triangle$  : 3-4 felt there was no glossiness.
  - imes: 2 or fewer felt there was no glossiness.

[0217]

25 (5) No stickiness

② : 8 or more felt there was no stickiness.

 $\bigcirc$  : 5-7 felt there was no stickiness.

 $\triangle$  : 3-4 felt there was no stickiness.

X : 2 or fewer felt there was no stickiness.

5 [0218]

(6) No discomfort

② : 8 or more felt there was no discomfort.

 $\bigcirc$  : 5-7 felt there was no discomfort.

 $\triangle$  : 3-4 felt there was no discomfort.

10  $\times$  : 2 or fewer felt there was no discomfort.

[0219]

[Examples 2-1 to 2-8, Comparative examples 2-1 to 2-11]

Skin treatment compositions for wrinkle

reduction were prepared by mixing ingredients with blend ratios (total blend ratio 100 wt%) according to the recipes shown in Tables 2-1 to 2-4. Also, the efficacy test was conducted on the skin treatment compositions for wrinkle reduction from the aforementioned Examples 2-1 to 2-8 and

Comparative examples 2-1 to 2-11; the evaluation results are also shown in Tables 2-1 to 2-4.

[0220] [Table 2-1]

	Example				
	2-1	2-2	2-3	2-4	
Ion-exchanged water	Balance	Balance	Ba l ance	Balance	
Glycerin	3. 0	3. 0	3. 0	3. 0	
Water dispersion of polyurethane of	0. 25	2. 5	_	12.5	
preparation example 1	(0.05)	(0.5)		(2.5)	
Water dispersion of polyurethane of	0. 25	2. 5	10. 0	5.0	
preparation example 2	(0. 05)	(0. 5)	(2. 0)	(1.0)	
Swollen form of non-emulsifying	_	40. 0	-	24. 0	
cross-linked silicone (Note 1)		(5. 0)		(3.0)	
Swollen form of non-emulsifying	20. 0	-	5. 0	-	
cross-linked silicone (Note 2)	(1.0)		(0. 25)		
Dimethyl silicone (20 mPa·s)	1. 0	1.0	1.0	1.0	
Sorbitan polyoxyethylene (10)	0. 5	0. 5	0. 5	0. 5	
monooleate (Note 6)					
Water dispersion of cross-linked	30. 0	30. 0	30. 0	30. 0	
sodium poly- $\gamma$ -glutamate (Note 8)	(0. 6)	(0. 6)	(0. 6)	(0. 6)	
Paraben	0. 15	0. 15	0. 15	0. 15	
Total	100	100	100	100	
Wrinkle reduction effect right after	0	0	0	0	
application					
Wrinkle reduction effect five hours	0	0	0	0	
after application					
Peeling	0	0	0	0	
Glossiness	0	0	0	0	
No stickiness	0	0	0	0	
No discomfort	0	0	0	0	

(Numbers in parentheses indicate the solid content.)

[0221] [Table 2-2]

	Example				
	2-5	2-6	2-7	2-8	
Ion-exchanged water	Balance	Balance	Balance	Balance	
Glycerin	3. 0	3. 0	3. 0	3. 0	
Water dispersion of polyurethane of	25. 0	35. 0	20. 0	30.0	
preparation example 1	(5. 0)	(7. 0)	(4.0)	(6.0)	
Water dispersion of polyurethane of	_	_	20. 0	20. 0	
preparation example 2			.(4. 0)	(4. 0)	
Swollen form of non-emulsifying	10. 0	_	-	40. 0	
cross-linked silicone (Note 1)	(1. 25)			(5. 0)	
Swollen form of non-emulsifying	_	30. 0	20. 0	-	
cross-linked silicone (Note 2)		(1.5)	(1.0)		
Dimethyl silicone (20 mPa·s)	1.0	1.0	1.0	1.0	
Sorbitan polyoxyethylene (10)	0. 5	0. 5	0. 5	0.5	
monooleate (Note 6)					
Water dispersion of cross-linked	30. 0	30. 0	30. 0	30. 0	
sodium poly- $\gamma$ -glutamate (Note 8)	(0. 6)	(0. 6)	(0. 6)	(0. 6)	
Paraben	0. 15	0. 15	0. 15	0. 15	
Total	100	100	100	100	
Wrinkle reduction effect right after	0	0	0	0	
application		:			
Wrinkle reduction effect five hours	0	0	0	0	
after application					
Peeling	0	0	0	0	
Glossiness	0	0	0	0	
No stickiness	0	0	0	0	
No discomfort	0	0	0	0	

(Numbers in parentheses indicate the solid content.)

[0222] [Table 2-3]

	Comparative example					
	2-1	2-2	2-3	2-4	2-5	2-6
Ion-exchanged water	Balance	Balance	Balance	Balance	Balance	Balance
Glycerin	3.0	3. 0	3. 0	3. 0	3.0	3. 0
Water dispersion of polyurethane of	2. 5	20. 0	_	-	_	35. 0
preparation example 1	(0.5)	(4. 0)				(7. 0)
Water dispersion of polyurethane of	2. 5	20. 0	-	-	-	_
preparation example 2	(0.5)	(4. 0)				
Swollen form of non-emulsifying	-	-	10.0	_	40.0	-
cross-linked silicone (Note 1)			(1. 25)		(5. 0)	
Dimethyl silicone (20 mPa·s)	1.0	1.0	1.0	1.0	1.0	1.0
Sorbitan polyoxyethylene (10)	0.5	0. 5	0. 5	0.5	0. 5	0. 5
monooleate (Note 6)						
Water dispersion of cross-linked	30. 0	30. 0	30. 0	30. 0	30. 0	30. 0
sodium poly-γ-glutamate (Note 8)	(0.6)	(0. 6)	(0. 6)	(0.6)	(0. 6)	(0.6)
Paraben	0. 15	0.15	0. 15	0. 15	0. 15	0. 15
Total	100	100	100	100	100	100
Wrinkle reduction effect right after application	×	0	Δ	×	0	0
Wrinkle reduction effect five hours after application	×	0	Δ	×	0	Δ
Peeling	×	×	0	×	×	×
Glossiness	Δ	×	Δ	0	0	Δ
No stickiness	0	×	×	0	×	×
No discomfort	0	×	0	×	Δ	×

(Numbers in parentheses indicate the solid content.)

[0223]

## [Table 2-4]

	Comparative example					
	2-7	2-8	2-9	2-10	2-11	
Ion-exchanged water	Balance	Balance	Balance	Balance	Balance	
Glycerin	3. 0	3. 0	3. 0	3. 0	3. 0	
Water dispersion of polyurethane of	10.0	-	_	-	_	
preparation example 1	(2. 0)					
Swollen form of non-emulsifying	_	_	40.0	-		
cross-linked silicone (Note 1)			(5. 0)			
Swollen form of non-emulsifying	-	_	_	10. 0	_	
cross-linked silicone (Note 2)	i			(0.5)		
Water dispersion of polyurethane	-	-	6. 06	_	-	
(Note 3)			(2.0)			
Dimethyl silicone (20 mPa·s)	-	_	_	_	10. 0	
Water dispersion of polyurethane	-	-	_	25. 0	_	
(Note 4)				(5. 0)		
Water dispersion of polyurethane	_	-	-	-	15. 0	
(Note 5)					(3.0)	
Sorbitan polyoxyethylene (10)	0. 5	0. 5	0. 5	0. 5	0. 5	
monooleate (Note 6)						
Trimethylsiloxysilicic acid (Note 7)	5. 0	-	-	-	-	
	(2. 5)					
Polyvinyl alcohol	_	3. 0	_	-	-	
Water dispersion of cross-linked	30. 0	30. 0	30. 0	30. 0	30. 0	
sodium poly- $\gamma$ -glutamate (Note 8)	(0. 6)	(0.6)	(0. 6)	(0. 6)	(0.6)	
Paraben	0. 15	0. 15	0. 15	0. 15	0. 15	
Total	100	100	100	100	100	
Wrinkle reduction effect right after	Δ	×	×	Δ	Δ	
application						
Wrinkle reduction effect five hours	×	×	Δ	×	Δ	
after application				_		
Peeling	Δ	×	Δ	×	Δ	
Glossiness	×	0	×	×	Δ	
No stickiness	0	×	0	×	×	
No discomfort	×	0	Δ	×	Δ	

(Numbers in parentheses indicate the solid

content.)

## 5 [0224]

In Tables 2-1 to 2-4:

- (Note 1) DC9045 (solid content 12.5%) (from Dow Corning Toray)
- (Note 2) KSG-44 (solid content approximately 5%) (from Shin-Etsu Chemical Co., Ltd.)
- 5 (Note 3) Avalure UR405 (solid content 33%) (from NOVEON)
  - (Average particle size: 100 nm, strength: 40  $\,$  kg/cm², elongation: 150%, film shrinkage rate: 35%)
- 10 (Note 4) Avalure UR445 (solid content 20%) (from NOVEON)
  - (Average particle size: 5 nm, strength: 240  $kg/cm^2$ , elongation: 610%, film shrinkage rate: 27%)
- 15 (Note 5) Iodosol PUD (solid content 20%) (from Nippon NSC)
  - (Average particle size: 20 nm, strength: 200 kg/cm $^2$ , elongation: 530%, film shrinkage rate: 23%)
- (Note 6) NIKKOL TO-10 (from Nikko Chemicals Co.) (Note 7) Contains KF7312J (decamethylcyclopentasiloxane solution; effective ingredient content 50%) (from Shin-Etsu Chemical Co., Ltd.).
- 25 (Note 8) Gelprotein A-8001 (solid content 2 %)

(from Idemitsu Technofine Co., Ltd.)
[0225]

As clearly shown in Tables 2-1 to 2-4, the skin treatment compositions for wrinkle reduction

5 from Examples 2-1 to 2-8 pertaining to the present invention are superior in terms of the wrinkle reduction effect and free of peeling, glossiness, stickiness, and discomfort. On the contrary, skin treatment compositions for wrinkle reduction from

10 Comparative examples 2-1 to 2-11 which do not meet the constituent requirements of the present invention, do not manifest the effects of the present invention.

[0226]

More Examples of the skin treatment composition for wrinkle reduction of the present invention are described below. The aforementioned efficacy test was conducted on these and the results were superior for all of them.

20 [0227]

[Example 2-9] Skin treatment composition for wrinkle reduction (cream type)

Ingredients

Blend ratio (wt%)

(1) Stearyl alcohol

6.0

25 (2) Stearic acid

2.0

(3)	Hydrated lanolin	4.0
(4)	Squalane	9.0
(5)	5-octyldodecanol	10.0
(6)	1,3-butylene glycol	6.0
(7)	Water dispersion of polyurethane	from
prep	aration example 1	5.0
	(soli	d content 1.0)
(8)	Water dispersion of polyurethane	from
prep	aration example 2	10.0
	(soli	d content 2)
(9)	Swollen form of non-emulsifying	cross-linked
sili	cone (Note 1)	20.0
	(soli	d content 3.2)
(10)	PEG 1500	4.0
(11)	Polyoxyethylene (20) sorbitan m	onococoate
(Not	e 2)	
		3.0
(12)	Glycerin monostearate	2.0
(13)	Ethylparaben	0.1
(14)	Butylparaben	0.1
(15)	Tocopherol	0.01
(16)	Perfume	0.1
(17)	Purified water	Balance
		Total 100.0
	(4) (5) (6) (7) prep (8) prep (9) sili (10) (11) (Not (12) (13) (14) (15) (16)	<pre>(4) Squalane (5) 5-octyldodecanol (6) 1,3-butylene glycol (7) Water dispersion of polyurethane preparation example 1</pre>

[0228]

(Note 1) DC9041 (solid content 16%) (from Dow Corning Toray)

(Note 2) NIKKOL TL-10 (from Nikko Chemicals Co.)
[0229]

- 5 (Preparation method)
  - (6), (10), (13), and (14) were added to (17) and the temperature was raised and adjusted to  $70^{\circ}$ C. Next, the oil phase consisting of (1), (2), (3), (4), (5), (9), (11), (12), (15), and (16)
- were prepared and the temperature was adjusted to 70℃. This was added to the water phase previously prepared; a homomixer was used to homogenize the emulsified particles and (7) and (8) were added. After deaeration, filtration, and
- 15 cooling, the target cream for wrinkle reduction was obtained.

[0230]

[Example 2-10] Skin treatment composition for wrinkle reduction (lotion type)

- 20 Ingredients Blend ratio (wt%)
  - (1) Palmitic acid 2.0
  - (2) Cetyl alcohol 1.5
  - (3) Petrolatum 4.0
  - (4) Squalane 5.0
- 25 (5) Glycerol tri-2-ethylhexanoate 2.0

	(6) Sorbitan monooleate	2.0
	(7) Dipropylene glycol	5.0
	(8) PEG 1500	3.0
	(9) Water dispersion of polyuretha	ane from
5	preparation example 1	2.0
	(sc	olid content 0.4)
	(10) Water dispersion of polyureth	nane from
	preparation example 2	8.0
	(sc	olid content 1.6)
10	(11) Swollen form of non-emulsify	ing cross-linked
	silicone (Note 1)	30.0
	(sc	olid content 1.8)
	(12) Triethanolamine	1.0
	(13) Methylparaben	0.1
15	(14) Phenoxy ethanol	0.1
	(15) Perfume	0.1
	(16) Purified water	Balance
		Total 100.0
	[0231]	
20	(Note 1) GRANSIL GCM-5 (solid con	tent
	approximately 6%) (from GRANT)	
	[0232]	
	<preparation method=""></preparation>	
	(7), (8), (13), and (14) wer	e added to (16)
25	and the temperature was raised and	d adjusted to

70%. The oil phase consisting of (1), (2), (3), (4), (5), (6), (11), and (15) were prepared and the temperature was adjusted to 70%. This oil phase was added to the water phase prepared

5 previously and pre-emulsification was carried out.

Next, (12) was added; after the emulsified

particles were homogenized with a homomixer, (9)

and (10) were added to obtain the target lotion

type for wrinkle reduction.

### 10 [0233]

[Example 2-11] Skin treatment composition for wrinkle reduction (cream type)

	Ingr	edients	Blend	ratio (wt	.%)
	(1)	Behenyl alcohol		1.0	
15	(2)	Batyl alcohol		2.0	
	(3)	Hydrated polyisobutene		4.0	
	(4)	Liquid petrolatum		9.0	
	(5)	Decamethylcyclopentasiloxan	n e	10.0	
20	(6)	1,3-butylene glycol		3.0	
	(7)	Glycerin		5.0	
	(8)	Water dispersion of polyure	ethane	from	
	prep	paration example 1		5.0	
			(solid	content	1.0)

(9) Water dispersion of polyurethane from2.5 preparation example 210.0

- (solid content 2.0)
- (10) Swollen form of non-emulsifying cross-linked silicone (Note 1) 20.0
  - (solid content 3.0)

5 (11) PEG 20000

- 4.0
- (12) Self emulsified glycerin monostearate (Note
- 2)

3.0

(13) Glycerin monostearate

2.0

(14) Ethylparaben

0.1

10 (15) Butylparaben

0.1

(16) Tocopherol

0.01

(17) Perfume

0.1

(18) Purified water

Balance

Total 100.0

**15** [0234]

<Preparation method>

- (6), (7), (11), (14), and (15) were added to (18) and the temperature was raised and adjusted
- to  $70^{\circ}$ C. Next, the oil phase consisting of (1),
- 20 (2), (3), (4), (5), (10), (12), (15), (16), and
  - (17) were prepared and the temperature was

    - phase previously prepared; a homomixer was used to
- homogenize the emulsified particles and (8) and
- 25 (9) were added. After deaeration, filtration, and

cooling, the target cream for wrinkle reduction
was obtained.

[0235]

(Note 1) GRANSIL ININ (solid content approximately
15%) (from GRANT)

(Note 2) NIKKOL MGS-ASE (from Nikko Chemicals Co.)

[0236]

[Example 2-12] Skin treatment composition for
wrinkle reduction (lotion type)

Ingredients

Blend ratio (wt%)

5

(4) Squalane

10	Ingredients	Blend ratio (wt%)
	(1) Palmitic acid	2.0
	(2) Cetyl alcohol	1.5
	(3) Petrolatum	4.0

- 15 (5) Glycerol tri-2-ethylhexanoate 2.0
  - (6) Sorbitan monooleate(7) Dipropylene glycol5.0
  - (8) PEG 1500 3.0
  - (9) Water dispersion of polyurethane from
- 20 preparation example 1 2.0
  - (10) Water dispersion of polyurethane from preparation example 2 8.0
    - (solid content 1.6)

(solid content 0.4)

5.0

25 (11) Swollen form of non-emulsifying cross-linked

silicone (Note 1)

30.0

(solid content 1.5)

(12) Triethanolamine

1.0

(13) Methylparaben

0.1

5 (14) Phenoxy ethanol

0.1

(15) Perfume

0.1

(16) Purified water

Balance

Total 100.0

[0237]

10 (Note 1) KSG-44 (solid content approximately 5%)

(from Shin-Etsu Chemical Co., Ltd.)

[0238]

<Preparation method>

- (7), (8), (13), and (14) were added to (16)
  15 and the temperature was raised and adjusted to
  70℃. The oil phase consisting of (1), (2), (3),
  (4), (5), (6), (11), and (15) were prepared and
  the temperature was adjusted to 70℃. This oil
  phase was added to the water phase prepared
- previously and pre-emulsification was carried out.

  Next, (12) was added; after the emulsified particles were homogenized with a homomixer, (9) and (10) were added to obtain the target lotion type for wrinkle reduction.
- **25** [0239]

```
[Example 2-13] Skin treatment composition for
    wrinkle reduction (gel type)
                                    Blend ratio (wt%)
    Ingredients
    (1) Copolymer of sodium polyacrylate/2-acrylamide-
    2-methylpropanesulfonic acid (AMPS) (Note 1)
5
                                                2.1
               (effective ingredient content 0.79)
    (2) Water dispersion of polyurethane from
                                                2.0
    preparation example 1
                                    (solid content 0.4)
10
    (3) Water dispersion of polyurethane from
                                                8.0
    preparation example 2
                                    (solid content 1.6)
    (4) Swollen form of non-emulsifying cross-linked
                                               30.0
15
    silicone (Note 2)
                                    (solid content 1.5)
    (5) Triethanolamine
                                                1.0
                                                0.1
    (6) Methylparaben
    (7) Phenoxy ethanol
                                                0.1
20
    (8) Perfume
                                                0.1
    (9) Purified water
                                               Balance
                                                5.0
    (10) Glycerin
                                                3.0
    (11) 1,3-butylene glycol.
                                          Total 100.0
```

105

[0240]

25

(Note 1) SIMULGE EG (effective ingredient content 37.5%) (from Sepic)

(Note 2) KSG-15 (solid content approximately 5%) (from Shin-Etsu Chemical Co., Ltd.)

5 [0241]

<Preparation method>

- (1), (5), (6), (7), (10), and (11) were added to (9), to which a mixture of (4) and (8) were added and dispersed homogeneously with a
- 10 homomixer. (2) and (3) were then added to obtain the target gel for wrinkle reduction.

[0242]

[Example 2-14] Skin treatment composition for wrinkle reduction (cream type)

15 Ingredients Blend ratio (wt%)

- (1) Behenyl alcohol 2.0
  - (2) Stearyl alcohol 3.0
  - (3) Octyl palmitate 3.0
- (4)  $\alpha$  -olefin oligomer 5.0
- 20 (5) Dimethylpolysiloxane (6 mPa·s) 10.0
  - (6) 1,3-butylene glycol 3.0
  - (7) Glycerin 5.0
  - (8) Water dispersion of polyurethane from preparation example 1 5
- 25 (solid content 1)

```
10.0
    preparation example 2
                                    (solid content 2.0)
    (10) Swollen form of non-emulsifying cross-linked
    silicone (Note 1)
                                               20.0
5
                                     (solid content 5)
    (11) PEG 20000
                                                4.0
    (12) Polyoxyethylene (20) sorbitan monococoate
    (Note 2)
                                                3.0
10
    (13) Glycerin monostearate
                                                2.0
                                                0.1
    (14) Ethylparaben
                                                0.1
    (15) Butylparaben
    (16) Tocopherol
                                                0.01
15
    (17) Perfume
                                                0.1
                                               Balance
    (18) Purified water
                                          Total 100.0
    [0243]
    (Note 1) KSG-16 (solid content approximately 25%)
    (from Shin-Etsu Chemical Co., Ltd.)
20
    (Note 2) NIKKOL TL-10 (from Nikko Chemicals Co.)
    [0244]
    <Preparation method>
         (6), (7), (11), (14), and (15) were added to
25
    (18) and the temperature was raised and adjusted
```

(9) Water dispersion of polyurethane from

to 70°C. Next, the oil phase consisting of (1), (2), (3), (4), (5), (10), (12), (15), (16), and (17) were prepared and the temperature was adjusted to 70°C. This was added to the water phase previously prepared; a homomixer was used to homogenize the emulsified particles and (8) and (9) were added. After deaeration, filtration, and cooling, the target cream for wrinkle reduction was obtained.